Section I—Air and Fallout

GROSS BETA ACTIVITY IN AIRBORNE PARTICULATES AND PRECIPITATION

Continuous surveillance of gross beta activity in air and precipitation provides one of the earliest and most sensitive indications of changes in environmental fission product activity from fallout. Although this surveillance does not provide enough information to assess human radiation exposure, it is used as an alerting system for determining when to intensify monitoring in other phases of the environment.

Surveillance data from a number of national programs are published monthly and summarized periodically to show current and long-range trends of atmospheric radioactivity in the Western Hemisphere. Data provided by programs of the Public Health Service, the Canadian Department of National Health and Welfare, the Mexican Commission of Nuclear Energy, and the Pan American Health Organization are presented individually in tabular form.

1. Radiation Surveillance Network July 1965

Division of Radiological Health
Public Health Service

Surveillance of atmospheric radioactivity in the United States is conducted by the Radiation Surveillance Network (RSN) of the PHS Division of Radiological Health which regularly gathers samples from 74 stations distributed throughout the country (figure 1). Most of the stations are operated by State health department personnel.

The alerting function of the network is provided by field estimates of the gross beta activity of airborne particulates on the filters. These determinations are performed about 5 hours after the sampling period to allow for decay of naturally-occurring radon daughters. The network station operators report (by telephone) field estimates greater than 10 pCi/m^3 (except for Hawaii, Alaska, and Puerto Rico, which report 5 pCi/m^3) to the Radiation Surveillance Center, Division of Radiological Health, Washington, D.C. Field estimates and laboratory measurements of daily concentrations and the estimated ages for selected samples are reported in the monthly RSN report (*1*). When unusually high air levels are reported, appropriate Federal and State officials are promptly notified.

Sampling and analytical procedures

Airborne particulates are collected continuously on carbon-loaded cellulose dust filters 4 inches in diameter. About 1,800 cubic meters of air are drawn through a filter during the 24-hour sampling period. The filters are forwarded to the RSN laboratory in Rockville, Maryland, where the gross beta activity is measured 4 days after the sampling period and again 7 days later if the net count is 2,000 cpm or higher. By using the two counts and the Way-Wigner formula (*2*), the age of fission products is estimated and the activity at time of collection determined.[1]

The total precipitation is sampled continuously at most RSN stations on a daily basis,

[1] If a sample contains a mixture of fresh and old fission products, the age estimated by the Way-Wigner formula is some intermediate value; consequently the calculated age of the fresh component will be overestimated.

Figure 1. Radiation Surveillance Network Sampling Stations

using funnels with collection areas of 0.4 square meter. If the rainfall exceeds 0.2 inch, the precipitation sample is analyzed for gross beta activity (*1*). Additionally, analyses for specific radionuclides may be performed.

In the laboratory the gross beta activity in evaporated precipitation samples is calculated by the same method used for analyzing the air filters. Total deposition for the sample is determined by:

$$\text{Deposition } (\text{nCi/m}^2) = \frac{\text{concentration (pCi/liter)} \times \text{precipitation depth (mm)}}{1{,}000}$$

Selected air and precipitation samples are analyzed by gamma spectrometry to determine the presence of fresh fission products. The method discussed by Burrus (*3*) and Covell (*4*) was adapted for resolving the complex gamma spectra.

Results

Monthly values of gross beta activity in air, total deposition, and depth of precipitation are given in table 1 for July 1965. During this month, the RSN collected 1,946 air samples and 332 precipitation samples. Forty-seven air samples were analyzed by gamma spectrometry. Although trace quantities of ^{140}Ba–La (below 1 pCi/m^3) were observed, no traces of other fisson products were apparent.

Time profiles of gross beta activity in air for eight stations are shown in figure 2.

Table 1. Gross beta activity in surface air and precipitation, July 1965

Station location		Air surveillance						Precipitation measurements	
		Number of samples		Gross beta activity, pCi/m³			Last profile in *RHD*	Total depth (mm)	Total deposition [b] (nCi/m²)
		Air	Pptn	Maximum	Minimum	Average [a]			
Ala:	Montgomery	31	8	0.40	<0.10	<0.15	May 65	130.8	<26.2
Alaska:	Adak	31		0.14	<0.10	<0.10	Sep 65	NS	
	Anchorage	29	10	0.27	<0.10	<0.12	May 65	38.4	<7.8
	Attu	17		0.10	<0.10	<0.10	Oct 65	NS	
	Fairbanks	27	4	0.24	<0.10	<0.14	Jun 65	21.9	<5.2
	Juneau	20	8	0.22	<0.10	<0.11	Jul 65	83.2	<16.8
	Kodiak	5		0.10	<0.10	<0.10	Aug 65	NS	
	Nome						Feb 65		
	Point Barrow	31		0.28	<0.10	<0.11	Nov 65	NS	
	St. Paul	29		0.11	<0.10	<0.10	Mar 65	NS	
Ariz:	Phoenix	29		0.37	<0.10	<0.14	Jul 65	NS	
Ark:	Little Rock	29	7	0.27	<0.10	<0.17	Jun 65	87.4	<17.5
Calif:	Berkeley	18		0.35	<0.10	<0.14	Aug 65	NS	
	Los Angeles	30		0.29	<0.10	<0.14	Feb 65	NS	
C. Z:	Ancon	14		0.18	<0.10	<0.11	Aug 65	NS	
Colo:	Denver	28	4	0.46	<0.10	<0.18	Aug 65	36.5	<7.4
Conn:	Hartford	30	5	0.63	<0.10	0.26	Jul 65	27.0	<20.3
Del:	Dover	19		0.83	<0.10	<0.27	May 65	NS	
D. C:	Washington	28	7	0.72	<0.10	<0.25	Nov 65	100.5	<20.1
Fla:	Jacksonville	31	10	0.35	<0.10	<0.15	Jun 65	175.2	<35.1
	Miami	29	13	0.30	<0.10	<0.13	Jul 65	168.2	<33.7
Ga:	Atlanta	8		0.25	<0.10	<0.14	Apr 65	NS	
Guam:	Agana	29		0.24	<0.10	<0.11	Apr 65	NS	
Hawaii:	Honolulu	31	6	0.14	<0.10	<0.10	Oct 65	58.1	<12.1
Idaho:	Boise	31		0.48	<0.10	0.24	Oct 65	NS	
Ill:	Springfield	31	1	0.76	0.12	0.28	Nov 65	12.5	<2.5
Ind:	Indianapolis	23	6	0.59	0.16	0.29	Apr 65	42.6	<8.7
Iowa:	Iowa City	30	6	0.59	0.13	0.25	Aug 65	61.0	<12.1
Kans:	Topeka	30	4	0.59	<0.10	0.21	May 65	73.3	<14.8
Ky:	Frankfort	27	7	0.66	<0.10	0.29	Nov 65	50.0	<10.1
La:	New Orleans	31	13	0.34	<0.10	<0.13	Nov 65	150.1	<30.4
Maine:	Augusta	30	7	0.81	<0.10	<0.34	Mar 65	47.0	<9.4
	Presque Isle	27	4	0.68	<0.10	<0.26	Aug 65	65.8	<13.2
Md:	Baltimore	21	3	1.06	<0.10	0.28	Jul 65	20.1	<4.0
	Rockville	19		0.88	<0.10	<0.25	Oct 65	NS	
Mass:	Lawrence	30	8	0.67	<0.10	0.30	May 65	68.6	<16.9
	Winchester	28	5	0.60	<0.10	<0.26	Sep 65	24.8	<5.9
Mich:	Lansing	31	4	0.86	<0.10	0.31	Oct 65	9.6	<2.0
Minn:	Minneapolis	21	7	0.53	<0.10	0.21	Apr 65	98.4	<19.6
Miss:	Jackson	16	2	0.26	<0.10	<0.15	Dec 64	56.8	<11.4
Mo:	Jefferson City	31	9	0.47	<0.10	0.23	Apr 65	68.3	<13.8
Mont:	Helena	30	7	0.71	<0.10	0.30	Sep 65	17.6	<4.6
Nebr:	Lincoln	14	5	0.61	<0.10	<0.27	Mar 65	161.2	<32.3
Nev:	Las Vegas	29		0.50	<0.10	<0.20	Jun 65	NS	
N. H:	Concord	21		1.08	<0.10	0.38	Nov 65	NS	
N. J:	Trenton	30	4	0.77	<0.10	<0.27	Mar 65	26.6	<5.3
N. Mex:	Santa Fe	26	8	0.13	<0.10	<0.11	Sep 65	30.1	<6.1
N. Y:	Albany	21	7	0.57	<0.10	0.32	Apr 65	49.6	<9.9
	Buffalo	31		0.71	<0.10	0.29	Aug 65	NS	
	New York	29		0.56	<0.10	<0.24	Sep 65	NS	
N. C:	Gastonia	30	8	0.58	<0.10	<0.17	Aug 65	131.6	<26.5
N. Dak:	Bismarck	30	11	0.53	<0.10	<0.24	Nov 65	107.8	<21.6
Ohio:	Cincinnati	6		0.17	<0.10	<0.12	May 65	NS	
	Columbus	29	6	1.03	0.13	0.37	Mar 65	110.8	<22.1
	Painesville	24	4	0.89	<0.10	0.34	Jul 65	61.0	<12.2
Okla:	Oklahoma City	30	2	0.23	<0.10	<0.14	Oct 65	50.0	<10.0
	Ponca City	30	7	0.21	<0.10	<0.11	Jul 65	136.3	<27.6
Ore:	Portland	31	3	0.69	<0.10	<0.23	Mar 65	6.9	<1.4
Pa:	Harrisburg	30	1	0.63	<0.10	<0.24	Apr 65	6.0	<1.2
P. R:	San Juan	31	4	0.25	<0.10	<0.12	Mar 65	93.0	<18.5
R. I:	Providence	30	5	0.73	<0.10	<0.29	Oct 65	48.4	<9.7
S. C:	Columbia	24	13	0.61	<0.10	<0.16	Sep 65	225.6	<45.2
S. Dak:	Pierre	31	3	0.62	<0.10	<0.24	Jul 65	19.9	<4.0
Tenn:	Nashville	30	7	0.49	<0.10	<0.17	Oct 65	66.5	<13.2
Tex:	Austin	28	1	0.24	<0.10	<0.14	May 65	8.8	<1.8
	El Paso	29	5	0.22	<0.10	<0.12	Nov 65	7.6	<1.8
Utah:	Salt Lake City	28	7	0.54	<0.10	<0.24	Feb 65	28.9	<5.9
Vt:	Barre	31	12	0.85	<0.10	0.34	Jun 65	73.1	<14.6
Va:	Richmond	31	9	0.68	<0.10	<0.20	Jun 65	194.7	<38.9
Wash:	Seattle	31	4	0.30	<0.10	<0.12	May 65	10.0	<2.0
	Spokane	30	2	0.54	<0.10	0.25	Apr 65	12.2	<2.6
W. Va:	Charleston	31	10	0.51	<0.10	<0.19	Oct 65	77.1	<15.4
Wis:	Madison	30	4	0.54	0.12	0.23	Jun 65	67.8	<13.5
Wyo:	Cheyenne	30	5	0.43	<0.10	<0.19	Jun 65	21.2	<6.0
Network summary [c]		1,946	332	1.08	<0.10	<0.20		67.2	<13.9

Key to symbols: Blank—no report received. NS—no sample collected.

[a] The monthly average is calculated by weighting the individual samples with the length of sampling period. Values of <0.10 are assumed to be 0.10 for averaging purposes. If the <0.10 values represent more than 10 percent of the values used in the average, a less-than sign is placed before the average.

[b] If any individual precipitation samples have activity concentrations under 200 pCi/liter, a less-than sign is placed before the total deposition value.

[c] For the network summary, all averages are arithmetic means of station averages.

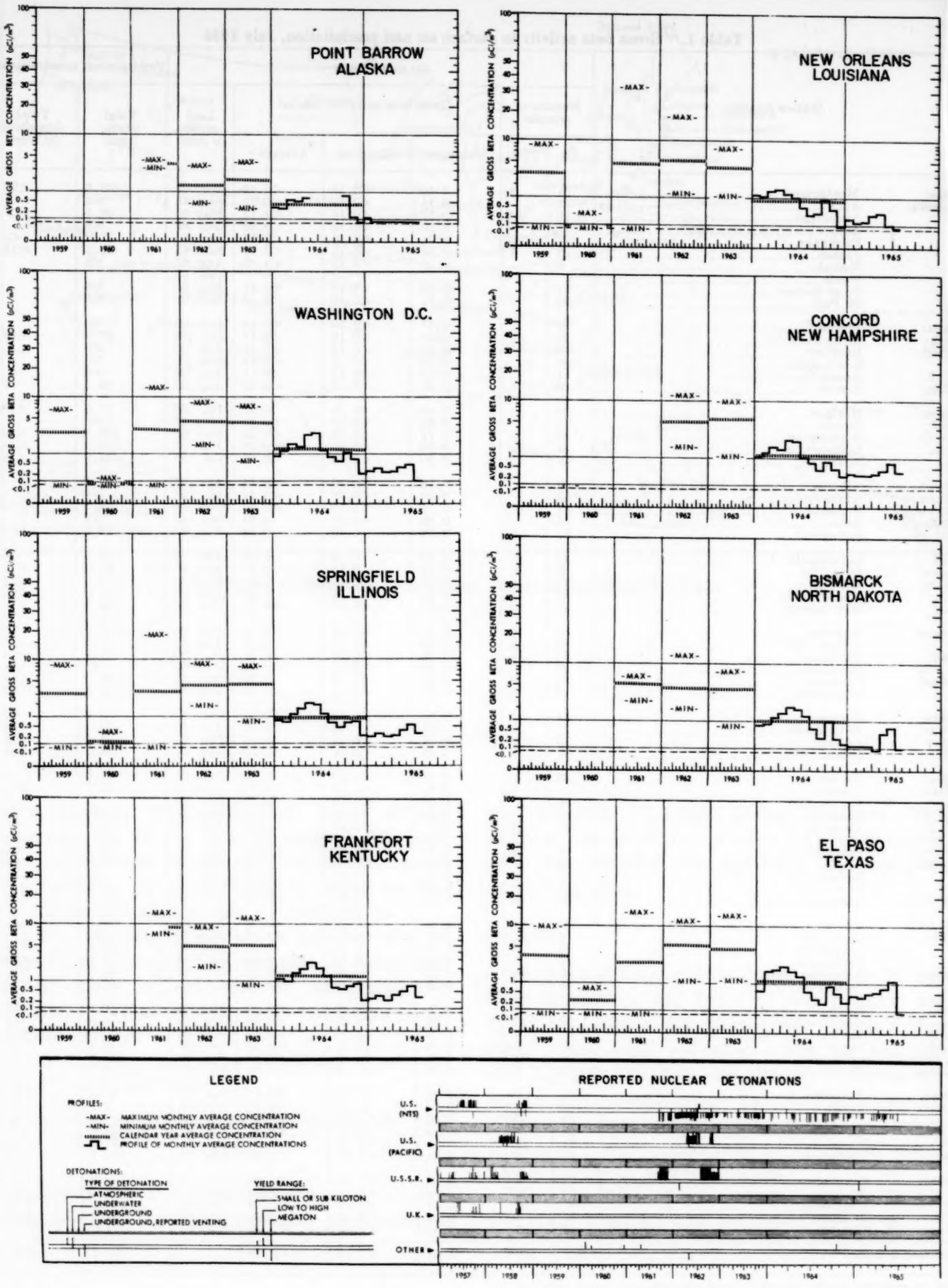

Figure 2. Monthly and yearly profiles of beta activity in air—Radiation Surveillance Network, 1959—July 1965

2. Canadian Air and Precipitation Monitoring Program, July 1965 [2]

Department of National Health and Welfare

The Radiation Protection Division of the Canadian Department of National Health and Welfare monitors air and precipitation in connection with its Radioactive Fallout Study Program. Twenty-four collection stations (figure 3) are located at airports, where the sampling equipment is operated by personnel from the Meteorological Services Branch of the Department of Transport. Detailed discussions of the sampling procedures, methods of analysis, and interpretation of results of the radioactive fallout program are contained in reports of the Department of National Health and Welfare (*5–10*).

Air sampling procedure and results

Each air sample involves the collection of particulates from about 650 cubic meters of air drawn through a high-efficiency 4-inch diameter glass-fiber filter during a 24–hour period. These filters are sent daily to the Radiation Protection Division Laboratory in Ottawa for analysis.

To determine the beta activity a 2-inch diameter disk is cut from each filter and counted with a thin-end-window, gas-flow, Geiger-Mueller counter system calibrated with a strontium–90—yttrium–90 standard. Four successive measurements are made on each filter to permit correction for natural activities and for the decay of short-lived fission products. The results are extrapolated to the end of the sampling period. Canadian air data for July 1965 are given in table 2.

Precipitation collection and analysis

The amount of radioactive fallout deposited on the ground is determined from measurements on material collected in special polyethylene-lined rainfall pots. The collection period for each sample is 1 month. After transfer of the water to the sample container, the polyethylene liner is removed, packed with the sample, and sent to the laboratory.

Strontium and cesium carriers are added to all samples on arrival at the laboratory. Other carriers are added to selected samples according to the specific radionuclides to be determined. The samples are then filtered and the filtrate evaporated to near dryness. The filter paper containing insoluble matter together with the polyethylene liner is then ignited and ashed at 450°C. The ash is combined with the soluble fraction, transferred to a glass planchet, evaporated under an infrared lamp, and then counted with a thin-end-window Geiger-

[2] Prepared from information and data in the August 1965 monthly report "Data from Radiation Protection Programs," Canadian Department of National Health and Welfare, Ottawa, Canada.

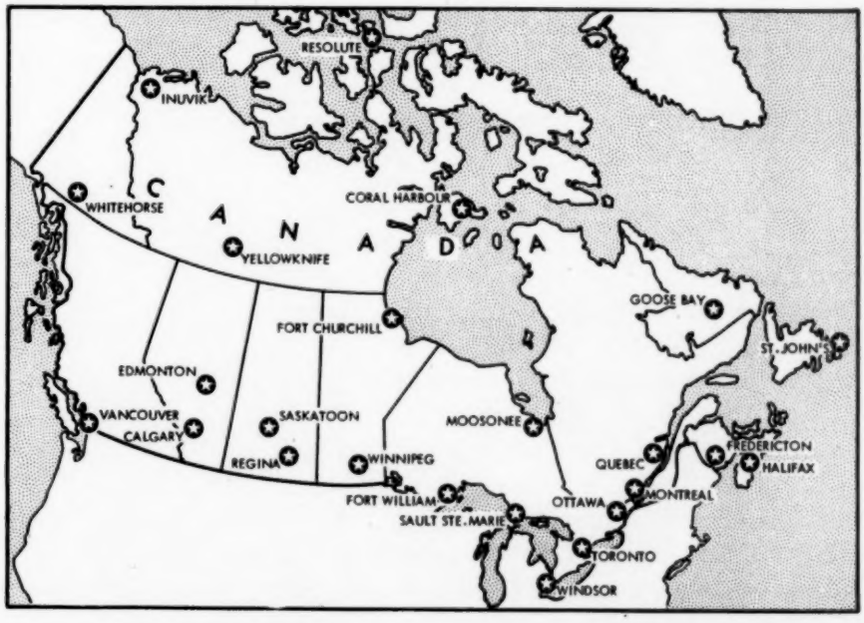

Figure 3. Canadian air and precipitation sampling stations

Mueller counter calibrated with a strontium–90—yttrium–90 source. Gross beta activities in precipitation for July 1965 are given in table 2.

The monthly precipitation samples represent the total deposition of radioactive materials on the earth's surface. The July 1965 gross beta deposition values are given in table 2. Results of radionuclide analyses of deposition are given in table 3. A review and discussion of fallout levels in Canada during 1964 have been presented by Booth *et al.* (*10*).

Table 2. Canadian gross beta activity in surface air and precipitation, July 1965

Station	Air surveillance				Precipitation measurements	
	Number of samples	Beta activity, pCi/m³			Average concentration (pCi/liter)	Total deposition (nCi/m²)
		Maximum	Minimum	Average		
Calgary	31	0.8	0.1	0.4	111	13.1
Coral Harbour	31	0.4	0.0	0.2	153	6.2
Edmonton	31	0.6	0.1	0.3	107	5.8
Ft. Churchill	31	0.6	0.0	0.2	114	4.7
Ft. William	31	0.8	0.0	0.3	124	5.2
Fredericton	31	0.8	0.1	0.3	117	5.3
Goose Bay	31	0.6	0.0	0.3	82	9.6
Halifax	31	0.7	0.0	0.3	78	4.3
Inuvik	31	1.0	0.0	0.3	96	4.4
Montreal	31	1.1	0.1	0.4	105	9.0
Moosonee	31	0.6	0.0	0.2	110	9.3
Ottawa	30	0.9	0.0	0.3	112	7.0
Quebec	30	1.1	0.0	0.3	135	14.6
Regina	31	0.6	0.2	0.3	175	7.5
Resolute	31	0.1	0.0	0.1	NS	NS
St. John's, Nfld	25	0.7	0.0	0.3	63	3.9
Saskatoon	31	0.4	0.1	0.2	448	13.9
Sault Ste. Marie	31	0.6	0.0	0.3	99	9.9
Toronto	31	0.8	0.0	0.4	106	10.3
Vancouver	31	0.3	0.0	0.1	278	1.6
Whitehorse	31	0.6	0.0	0.2	276	3.0
Windsor	31	1.0	0.1	0.3	79	5.5
Winnipeg	31	0.6	0.0	0.2	78	6.2
Yellowknife	2	0.2	0.2	0.2	158	5.8
Average		0.7	0.0	0.3	139	7.2

NS—no sample.

Table 3. Radionuclide deposition (nCi/m²) in Canadian fallout, July 1965[a]

Station	^{89}Sr	^{90}Sr	^{137}Cs	^{95}Zr	^{140}Ba
Calgary	0.70	1.17	1.69	0.16	0.28
Coral Harbour	0.26	0.28	0.44	NA	NA
Edmonton	0.00	0.78	1.14	NA	NA
Ft. Churchill	0.32	0.36	0.56	NA	NA
Ft. William	0.21	0.37	0.62	NA	NA
Fredericton	0.25	0.46	0.72	NA	NA
Goose Bay	0.55	0.92	1.17	NA	NA
Halifax	0.20	0.34	0.36	0.02	0.13
Inuvik	0.27	0.44	0.52	NA	NA
Montreal	0.59	0.74	1.06	0.21	0.46
Moosonee	0.73	0.68	1.00	NA	NA
Ottawa	0.28	0.68	0.99	NA	NA
Quebec	1.05	1.35	1.98	NA	NA
Regina	0.33	0.61	0.88	NA	NA
Resolute	NS	NS	NS	NS	NS
St. John's, Nfld	0.52	0.44	0.65	NA	NA
Saskatoon	0.38	0.76	1.56	NA	NA
Sault Ste. Marie	0.65	0.78	1.05	NA	NA
Toronto	0.56	0.81	0.91	NA	NA
Vancouver	0.00	0.04	0.11	0.02	0.07
Whitehorse	0.12	0.27	0.46	NA	NA
Windsor	0.28	0.36	0.60	NA	NA
Winnipeg	0.36	0.53	0.75	0.10	0.09
Yellowknife	0.33	0.53	0.63	NA	NA
Average	0.39	0.60	0.86	0.10	0.21

Key to symbols:
NA—no analysis.
NS—no sample.
[a] Samples from all stations are routinely analyzed for ^{89}Sr, ^{90}Sr, and ^{137}Cs; samples from five selected stations are regularly analyzed for ^{95}Zr and ^{140}Ba. Negligible quantities are not reported.

3. Mexican Air Monitoring Program July 1965

National Commission of Nuclear Energy

The Radiation Surveillance Network of Mexico was established by the Commision Nacional de Energía Nuclear (CNEN), Mexico City. From 1952 to 1961 the network was directed by the Institute of Physics of the University of Mexico, under contract to the CNEN (*11–15*).

In 1961, the CNEN appointed its Division of Radiological Protection (DRP) to establish a new radiation surveillance network. This network consists of 17 stations (see figure 4), 12 of

Figure 4. Fallout network sampling stations in Mexico

which are located at airports and operated by airline personnel. The remaining five stations are located at Mexico City, Mérida, Veracruz, San Luis Potosí, and Enseñada. Staff members of the DRP operate the station at Mexico City, while the other four stations are manned by members of the Centro de Previsión del Golfo de México, the Chemistry Department of the University of Mérida, the Instituto de Zonas Desérticas of the University of San Luis Potosí and the Escuela Superior de Ciencias Marinas of the University of Baja California, respectively.

Sampling

The sampling procedure involves drawing air for 24 hours a day, 3 to 4 days a week at the rate of approximately 1,200 cubic meters per day, through a high-efficiency, 6- x 8-inch glass fiber filter, using high-volume samplers. After each 24-hour sampling period, the filter is removed and forwarded via airmail to the "Laboratorio de Estudios sobre Contaminación Radiactiva," CNEN, in Mexico City for assay of gross beta activity. A minimum of 3 to 4 days after collection is allowed for decay of radon and thoron daughter natural radioactivity. Data are not extrapolated back to the date of collection.

Results

The maximum, minimum, and average fisson product beta concentrations in surface air during July 1965 are presented in table 4.

Table 4. Gross beta activity of airborne particulates, Mexico, July 1965

Station	Number of samples	Gross beta activity, pCi/m³		
		Maximum	Minimum	Average
Acapulco	16	<0.1	<0.1	<0.1
Ciudad Juárez	15	0.2	<0.1	0.1
Chihuahua	20	0.3	<0.1	0.1
Enseñada	3	0.1	0.1	0.1
Guadalajara	13	<0.1	<0.1	<0.1
Guaymas	6	<0.1	<0.1	<0.1
La Paz	16	0.9	<0.1	0.2
Matamoros	5	0.2	<0.1	0.1
Mazatlán	16	0.3	<0.1	0.1
Mérida	22	0.1	<0.1	<0.1
México, D.F.	8	0.1	<0.1	<0.1
Nuevo Laredo	8	0.3	<0.1	0.1
San Luis Potosí	15	0.1	<0.1	<0.1
Tampico	17	0.1	<0.1	<0.1
Torréon	28	0.4	<0.1	0.1
Tuxtla Gutiérrez	NS	NS	NS	NS
Veracruz	14	0.2	<0.1	<0.1

NS, no sample collected.

4. Pan American Air Sampling Program July 1965

Pan American Health Organization and Public Health Service

Gross beta activity in air is monitored by six countries in the Americas under the auspices of a collaborative program developed by the Pan American Health Organization and the Public Health Service (PHS), for assisting countries of the Americas in developing radiological health programs. The sampling equipment and analytical services are provided by the Division of Radiological Health, PHS, and are identical with those employed for the Radiation Surveillance Network.

The six air sampling stations included in the Program are operated by the technical staff of the Ministry of Health in each country. The station in Kingston, Jamaica, is operated by the Public General Hospital; in Caracas, Venezuela, by the Venezuelan Institute for Scientific Investigations; in Lima, Peru, by the Institute of Occupational Health; in Santiago, Chile, by the Occupational Health Service; in Trinidad, West Indies, by the University of the West Indies; and in Buenos Aires, Argentina, by the Ministry of Social Welfare and Public Health.

The May 1965 air monitoring results from the six participating countries are given in table 5.

Table 5. Gross beta activity in surface air (concentrations in pCi/m³), July 1965

Sampling stations	Number of samples	Maximum	Minimum	Average [a]
Kingston, Jamaica	14	0.22	<0.10	<0.12
Caracas, Venezuela	17	0.15	<0.10	<0.10
Lima, Peru	24	0.10	<0.10	<0.10
Santiago, Chile	16	0.19	<0.10	<0.12
Trinidad, West Indies	14	0.19	<0.10	<0.11
Buenos Aires, Argentina	NS	NS	NS	NS

[a] The monthly average is calculated by weighting the individual samples with length of sampling period. Values of <0.10 are assumed to be 0.10 for averaging purposes. If the <0.10 values represent more than 10 percent of the values used in the average, a less-than sign is placed in front of the average.

NS, no samples analyzed by laboratory.

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RADIOCARBON IN SURFACE AIR

J. C. Drobinski, Jr.[1]

Prior to 1950, the natural radioactive carbon dioxide content of the troposphere and lower stratosphere was in a steady-state equilibrium (*1*); that is, conditions were such that the cosmic ray production of carbon–14 by the (n, p) reaction on nitrogen in the upper atmosphere equalled its radioactive decay. Under these conditions, the natural level of carbon–14 in the atmosphere and biosphere was about 14 ± 1 disintegrations per minute per gram of carbon. Recently, it has been broadly established that this equilibrium has existed, probably with minor fluctuations, for at least the last 300,000 years (*2*).

The detonation of fusion and high-yield fission devices, which began in 1952, has added macro amounts of carbon–14 to the natural reservoir. It appears that most of this activity was injected into the stratosphere followed by only a gradual entrance into the troposphere. As a result, the rise in tropospheric levels of carbon–14 has been slow. Data on carbon–14 concentrations in the stratosphere have been reported by Hagemann (*3*). Table 1 shows increases in the carbon–14 levels reported for 1956–1962 in the northern troposphere (*4*).

Table 1. Tropospheric inventory of artificial carbon-14

Year	Increase, percent [a]
1956	8
1957	12
1958	18
1959	21
1960	21
1961	24
1962	28

[a] Over 1950 level.

Although the dose rate from radioactive carbon–14 is very low, its long half-life (5,740 years) (*5*) and its ultimate entry into the biosphere make it probably the most significant radionuclide of genetic importance produced by nuclear detonations (*6,7*). Since very little carbon is absorbed through respiration, its principal route of entry into man is through the food chain. Carbon–14, as carbon dioxide, is incorporated into plant tissue through photosynthesis; it then enters the human food chain either directly or through animal intermediates. Because the carbon–14 content in animals tends to lag behind surface air levels (*8–11*), data on the concentrations of carbon–14 in surface air can serve as an early indicator of possible changes in the ecological system leading to man.

Sample collection

Carbon dioxide is collected at Northeastern Radiological Health Laboratory approximately every two weeks. The intake is located about 20 feet above ground level. A dehumidifier-compressor,[2] passes air through silica gel to remove moisture. The air then passes through a trap cooled with dry ice-ethanol (–78° C.) to remove any remaining traces of water vapor and then to a bed of Linde molecular sieve (type 5A) which absorbs the carbon dioxide, while permitting the oxygen, nitrogen, and inert gases to pass through. Prior removal of moisture is necessary since the molecular sieve also absorbs water. Although molecular sieves can be operated at high flow rates with good efficiency and low-pressure drop, the particular dehumidifier-compressor unit available has a maximum operating capacity of 40 cubic feet of air per hour. Therefore the dehumidifying-absorption process is continued for 24–26 hours to obtain sufficient sample (about 8 liters of carbon dioxide,[3] corresponding to 4.3 grams of carbon). Collection of the sample over such a long period also minimizes the effect of transient meterological conditions.

After the carbon dioxide has been absorbed by the molecular sieve at room temperature, it is transferred to a collection system. The carbon dioxide is desorbed from the sieve material at 500° C. and is collected in a trap cooled by liquid nitrogen (–196° C.) after passage through a trap cooled by dry ice-ethanol (–78° C.). Volatile impurities (air) are removed by pumping to a pressure of 15 microns while the carbon dioxide is frozen in the liquid nitrogen trap. Finally, the gas is sublimed into

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[2] Communication Products Company, Marlboro, New Jersey.

[3] At standard conditions of temperature and pressure.

storage tanks where the quantity collected is measured. Observed yields have been 95 percent or more.

Purification and counting

The carbon dioxide is then transferred to a purification system where it is purified by a series of vacuum evaporation-condensation steps through activated coconut charcoal. The final "evaporation" is carried out at –78° C. so that any radon present will be retained on the charcoal. In this "evaporation," the CO_2 also passes through phosphorus pentoxide (P_2O_5) before transfer to the counters.

The carbon–14 content is measured in gas-phase carbon dioxide proportional counters. The counting chamber volume is 7.5 liters, at an operating pressure of 30 cm. mercury. Background is reduced by an iron shield 12-inches thick and by an annular anti-coincidence shield surrounding the counter. The system has been described by Drobinski (*11*). Background is about 20 counts per minute and the standard deviation of the net activity is about 1 cpm for a 180-minute counting period.

Results and interpretation

Table 2 lists the carbon–14 activities observed in the atmosphere from January 1964 to April 1965. The actual activity (disintegrations per minute of carbon–14 per gram of carbon) and the percentage increase over 1950 levels (△carbon–14) are presented. The latter factor is calculated as follows:

$$\Delta\text{carbon-14} = \frac{(\text{dpm/g}) - 14}{14} \times 100$$

where 14 is the 1950 activity in dpm/g of carbon.

The data in the table show an increase in the carbon–14 content over the period of study, suggesting the entrance of this nuclide into the troposphere from the stratospheric inventory. Most of the increase was noted in March 1964. Subsequent to this date, the level remained fairly constant. Fluctuations in carbon–14 concentrations from sample to sample which are greater than analytical error may be due to meteorological conditions. The low values of some samples may be due to dilution with inactive carbon dioxide resulting from the combustion of fossil fuels (Suess effect) (*12*). Although some geographical variability of carbon–14 may be expected, the data obtained at Winchester agree in general with those reported elsewhere (*10, 13*).

Table 2. Carbon-14 content in surface air, Winchester, Massachusetts

Date of sampling	dpm/g C	Percent $\Delta^{14}C$ [a]	Date of sampling	dpm/g C	Percent $\Delta^{14}C$ [a]
1964			Sept 15	22.6±1.0	61
			Oct 15	22.5±1.0	60
Feb 5	17.0±1.2	21	Nov 16	23.7±1.0	69
Feb 10	19.2±1.1	37	Dec 1	24.8±1.0	77
Mar 3	22.0±0.9	57	Dec 15	23.9±1.5	70
Mar 17	21.9±0.8	57			
Mar 31	23.3±1.1	66	1965		
April 15	23.4±0.8	67			
April 28	20.1±1.0	43	Jan 5	21.2±1.1	52
May 14	21.8±0.9	56	Feb 2	19.0±1.0	36
June 3	22.6±1.0	61	Feb 17	22.1±1.0	58
June 23	21.9±1.0	56	Mar 2	21.3±1.0	52
July 15	22.9±1.0	63	Mar 16	24.2±1.1	73
Aug 3	23.3±1.0	67	Mar 31	21.0±1.0	50
Aug 14	22.3±1.0	59	April 13	21.7±1.0	55

[a] Percent above natural level of 14±1 dpm/g carbon in 1950.

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Section II—Milk and Food

MILK SURVEILLANCE

Although milk is only one of the many sources of dietary intake of radionuclides, it is the single food item most often used as an indicator of the population's intake of radionuclides from the environment. This is because fresh milk is consumed by a large segment of the United States population and contains most of the radionuclides occurring in the environment which have been identified as biologically important. In addition, milk is produced and consumed on a regular basis, is convenient to composite and analyze, and samples representative of milk consumption in any area can be readily obtained.

1. Pasteurized Milk Network, July 1965

Division of Radiological Health and Division of Environmental Engineering and Food Protection, Public Health Service

The Public Health Service pasteurized milk surveillance program had its origin in a raw milk monitoring network (*1*) established by the Service in 1957. One of the primary objectives of this network was the development of methods for milk collection and radiochemical analysis suitable for larger scale programs.

Experience derived from this study led to the activation of a pasteurized milk sampling program with stations selected to provide nationwide surveillance of milk production and consumption areas. The present Pasteurized Milk Network (PMN), which consists of 63 stations, has at least one station in every State, the Canal Zone, and Puerto Rico.

Sampling procedure

Samples are collected through the cooperation of State and local milk sanitation authorities. According to established procedures, the sample at each station is composited of subsamples from each milk processing plant in proportion to the plant's average sales in the community served. At most stations the sample represents from 80 to 100 percent of the milk processed. Prior to September 15, 1961, the composite sample was taken from 1 day's sales per month and was as representative of the community's supply as possible under practical conditions. Beginning with the resumption of nuclear weapons testing in the atmosphere in September 1961, and continuing through January 1963, samples were collected twice a week at nearly all stations and daily for short periods at selected stations. Since then the sampling frequency has been reduced to once a week.

Samples are preserved with formaldehyde and sent to the PHS Southwestern (SWRHL), Southeastern (SERHL) or Northeastern (NERHL) Radiological Health Laboratories for analysis. Gamma analyses for iodine–131 are made within 3 to 6 days after sample collection, and any results exceeding 100 pCi/liter are immediately telephoned to appropriate State health officials for possible public health action. Analytical results are normally available 6 to 7 weeks after weekly samples are received by the laboratories.

Analytical procedures

Iodine–131, cesium–137, and barium–140 concentrations are determined by gamma scintillation spectrometry.[1] After the weekly samples are gamma-scanned, samples from 2 consecutive weeks are composited and analyzed

[1] Southeastern Radiological Health Laboratory employs a radiochemical procedure for barium–140 analysis.

radiochemically to determine strontium–89 and strontium–90. With all such measurements there is an inherent statistical variation. For the low radionuclide levels usually observed in milk and other environmental samples, this variation on a percentage basis is relatively high. It depends upon such factors as the method of chemical analysis, the sample counting rate and counting time, interferences from other radionuclides, and the background count. For milk samples, counting times of 50 minutes for gamma spectrometry and 30 to 50 minutes for beta determinations are used. Table 1 shows the approximate analytical errors (including counting error) associated with determinations of radionuclide concentrations in milk. These errors were determined by comparing results of a large number of replicate analyses.

Table 1. Analytical errors associated with determinations of radionuclides concentrations in milk

Nuclide	Concentration (pCi/liter)	Error [a] (pCi/liter)	Concentration (pCi/liter)	Error [a] (percent of concentration)
Iodine-131	less than 100	10	100 or greater	10
Barium-140	less than 100	10	100 or greater	10
Cesium-137	less than 100	10	100 or greater	10
Strontium-89	less than 50	5	50 or greater	10
Strontium-90	less than 20	2	20 or greater	10

[a] Two standard deviations

The minimum detectable concentration is defined as the measured concentration equal to the two-standard deviation analytical error. Accordingly, the minimum detectable concentrations in units of pCi/liter are: strontium–89, 5; strontium–90, 2; cesium–137, 10; barium–140, 10; and iodine–131, 10. At these levels and below, the counting error comprises nearly all of the analytical error.

Calcium analyses at SERHL are done by an ion exchange and permanganate titration method, while at NERHL and SWRHL an ethylenediaminetetraacetic acid (EDTA) method is used. Stable potassium concentrations are estimated from the potassium–40 concentrations determined from the gamma spectrum, but are not reported herein.

Data presentation

Table 2 presents summaries of the analyses for July 1965. Radionuclide values reported by a laboratory as being below the minimum detectable concentration have been averaged by using one-half the minimum detectable value. A modified averaging procedure has been used for iodine–131 and barium–140 since October 1963, when nondetectable concentrations of these radionuclides began to be considered zero. A similar procedure is used for the network average.

Figures 1 and 2 are isogram maps showing the estimated strontium–90 and cesium–137 concentrations in milk over the entire country. The value printed beside each station is the monthly average concentration for that station. The isograms were developed by arbitrary interpolation between values for the individual stations.

The distribution of monthly average strontium–90 concentrations as observed in pasteurized milk for the months February through July 1965 is presented in table 3. The maximum monthly average concentration of strontium–90 for July was 37 pCi/liter, observed at Minot, North Dakota. This level is in Range II as defined by the Federal Radiation Council (20–200 pCi/liter). The overall PMN average monthly strontium–90 concentration in pasteurized milk for July was 17.2 pCi/liter as compared to 20.0 pCi/liter in June 1965. The average monthly strontium–90 concentrations in pasteurized milk from selected cities in the sampling program are presented in figure 3.

The network average cesium–137 concentration in milk for July 1965 was 51 pCi/liter as compared to 61 pCi/liter in June 1965. The maximum monthly average cesium–137 concentration in pasteurized milk was 185 pCi/liter, observed at Tampa, Florida. The distribution of monthly average cesium–137 concentrations in milk for the months February through July 1965 is presented in table 4.

The decrease in radionuclide concentrations in milk observed during February through July 1965 can be expected to continue in the absence of atmospheric nuclear testing.

During July 1965, four stations exhibited an iodine–131 monthly average of 10 pCi/liter. The presence of iodine–131 and strontium–89 in pasteurized milk during July 1965 is presumed a consequence of the mainland China atmospheric nuclear test of May 14, 1965.

Table 2. Average concentrations of stable elements and radionuclides in pasteurized milk, second quarter 1965 and July 1965[a]

Sampling locations		Calcium (g/liter)		Strontium-89 (pCi/liter)		Strontium-90 (pCi/liter)		Cesium-137 (pCi/liter)		Iodine-131 (pCi/liter)	
		Second quarter	Average for July	Second quarter	Average for July	Second quarter	Average for July	Second quarter	Average for July	Second quarter	Average for July
Ala:	Montgomery	1.17	1.12	<5	<5	18	13	55	40	0	0
Alaska:	Palmer	1.22	1.23	<5	<5	20	13	65	50	0	0
Ariz:	Phoenix	1.21	1.21	<5	<5	5	5	20	20	0	0
Ark:	Little Rock	1.15	1.13	5	15	38	31	70	60	10	0
Calif:	Sacramento	1.25	1.31	<5	<5	8	6	35	20	0	0
	San Francisco	1.26	1.27	<5	<5	11	7	35	20	0	0
C.Z:	Cristobal	1.10	1.11	<5	<5	4	4	35	30	0	0
Colo:	Denver	1.33	1.26	<5	10	19	15	70	45	0	0
Conn:	Hartford	1.11	1.09	<5	<5	15	15	75	60	0	0
Del:	Wilmington	1.14	1.12	<5	5	19	16	70	50	0	0
D. C:	Washington	1.15	1.12	5	<5	17	14	55	40	10	0
Fla:	Tampa	1.16	1.16	<5	<5	12	11	175	185	0	0
Ga:	Atlanta	1.15	1.14	<5	<5	29	21	85	60	0	0
Hawaii:	Honolulu	1.23	1.16	<5	<5	10	7	55	40	0	0
Idaho:	Idaho Falls	1.27	1.26	<5	<5	20	23	95	60	10	10
Ill:	Chicago	1.13	1.11	<5	<5	17	11	75	40	10	0
Ind:	Indianapolis	1.15	1.12	5	<5	19	14	55	30	10	0
Iowa:	Des Moines	1.26	1.18	10	5	23	20	55	35	20	0
Kans:	Wichita	1.25	1.20	<5	5	18	19	50	35	10	0
Ky:	Louisville	1.15	1.12	5	10	24	20	45	30	10	0
La:	New Orleans	1.20	1.16	5	5	42	34	80	60	10	0
Maine:	Portland	1.12	1.11	<5	10	21	23	125	110	0	0
Md:	Baltimore	1.14	1.10	5	<5	19	17	60	50	10	0
Mass:	Boston	1.13	1.11	<5	10	22	22	125	100	0	0
Mich:	Detroit	1.13	1.11	<5	<5	17	11	75	40	10	0
	Grand Rapids	1.16	1.14	<5	5	19	16	80	55	10	0
Minn:	Minneapolis	1.29	1.31	10	10	28	26	90	65	20	
Miss:	Jackson	1.18	1.18	5	<5	34	29	55	45	0	0
Mo:	Kansas City	1.25	1.22	10	10	22	19	45	30	30	0
	St. Louis	1.20	1.19	5	10	19	16	45	25	20	0
Mont:	Helena	1.28	1.23	<5	10	20	19	95	65	10	0
Nebr:	Omaha	1.19	1.21	5	<5	21	17	50	35	20	0
Nev:	Las Vegas	1.28	1.18	<5	<5	7	8	40	20	10	0
N. H:	Manchester	1.15	1.13	<5	5	25	25	150	130	0	0
N. J:	Trenton	1.11	1.09	<5	5	16	16	70	45	0	0
N. Mex:	Albuquerque	1.23	1.23	<5	<5	11	8	35	20	0	0
N. Y:	Buffalo	1.10	1.11	<5	5	17	12	95	55	0	0
	New York	1.11	1.09	<5	5	21	20	90	65	0	0
	Syracuse	1.10	1.11	<5	10	18	12	80	45	0	0
N. C:	Charlotte	1.20	1.12	5	<5	27	25	65	50	0	0
N. Dak:	Minot	1.25	1.27	10	20	57	37	120	70	10	0
Ohio:	Cincinnati	1.13	1.12	<5	5	18	14	50	30	10	0
	Cleveland	1.13	1.11	5	10	20	13	75	40	10	0
Okla:	Oklahoma City	1.12	1.08	5	5	20	16	45	30	10	0
Ore:	Portland	1.30	1.30	<5	<5	24	17	90	50	0	0
Pa:	Philadelphia	1.13	1.05	<5	<5	19	15	65	40	0	0
	Pittsburgh	1.14	1.12	5	10	25	22	95	65	10	0
P. R:	San Juan	1.12	1.11	<5	<5	11	10	50	45	0	0
R. I:	Providence	1.12	1.12	<5	<5	20	19	95	85	0	0
S. C:	Charleston	1.16	1.13	<5	<5	27	23	100	70	0	0
S. Dak:	Rapid City	1.05	1.04	10	20	33	28	105	55	10	10
Tenn:	Chattanooga	1.19	1.14	5	5	30	26	65	45	0	0
	Memphis	1.15	1.13	5	10	26	21	45	30	10	0
Tex:	Austin	1.12	1.12	<5	<5	8	6	25	25	0	10
	Dallas	1.17	1.11	5	<5	20	14	45	30	0	0
Utah:	Salt Lake City	1.39	1.34	<5	<5	26	15	120	80	10	0
Vt:	Burlington	1.12	1.11	<5	5	21	21	100	75	0	0
Va:	Norfolk	1.17	1.14	5	<5	21	18	60	45	10	10
Wash:	Seattle	1.27	1.26	<5	<5	25	19	105	80	0	0
	Spokane	1.30	1.30	<5	<5	27	23	90	60	0	0
W. Va:	Charleston	1.12	1.10	10	5	20	19	40	30	10	0
Wis:	Milwaukee	1.19	1.19	<5	5	17	11	85	45	10	0
Wyo:	Laramie	1.23	1.31	5	<5	17	17	70	55	10	0
Network average		1.18	1.16	<5	5	20.7	17.2	72	51	6	1

[a] Results of all barium-lanthanum-140 analyses were zero.

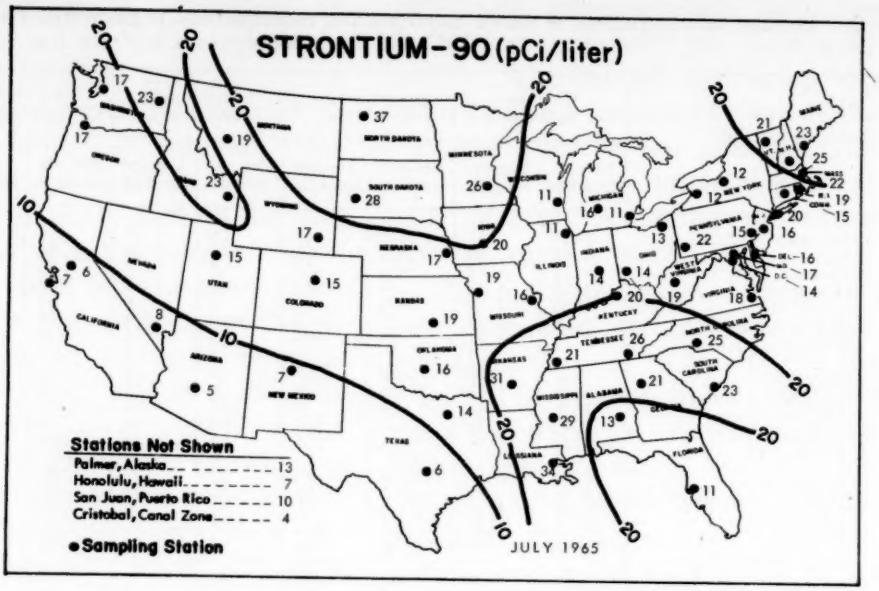

Figure 1. Strontium–90 concentrations in pasteurized milk, July 1965

Table 3. Frequency distribution, strontium-90 concentrations in milk at PMN stations, July 1964 and February–July 1965

Strontium-90 (pCi/liter)	Number of stations						
	1964	1965					
	July	Feb	Mar	Apr	May	June	July
Under 10	6	3	5	5	4	7	8
10–19	6	31	29	23	26	21	34
20–29	29	22	21	27	25	29	18
30–39	14	5	6	5	5	4	3
40–49	4	2	1	2	2	2	0
50–59	1	0	1	0	0	0	0
60–69	2	0	0	1	1	0	0
70–79	0	0	0	0	0	0	0

Table 4. Frequency distribution, cesium-137 concentrations in milk at PMN stations, July 1964 and February–July 1965

Cesium–137 (pCi/liter)	Number of stations						
	1964	1965					
	July	Feb	Mar	Apr	May	June	July
Under 50	4	8	9	7	17	19	34
50–99	24	38	36	35	31	38	25
100–149	21	15	16	18	13	5	3
150–199	8	2	2	3	2	1	1
200–249	3	0	0	0	0	0	0
250–299	2	0	0	0	0	0	0

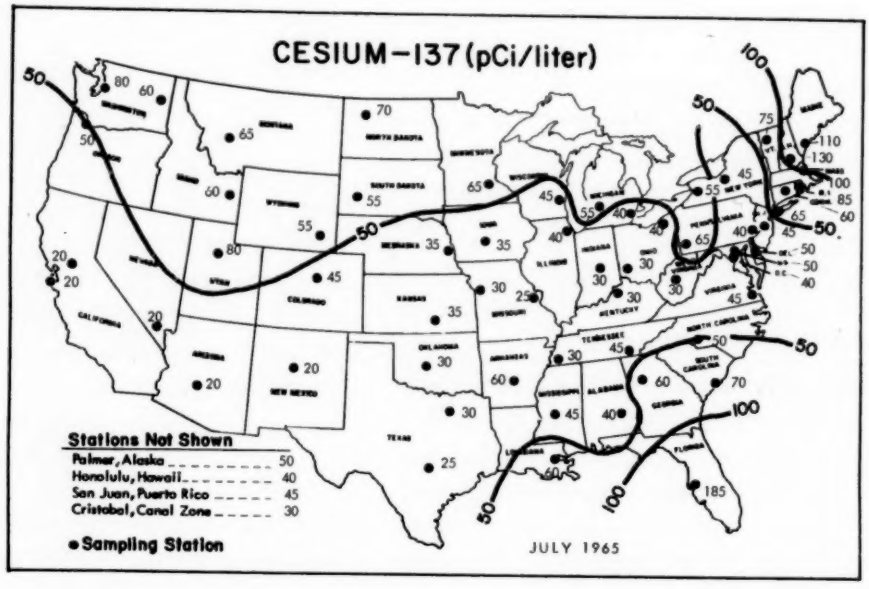

Figure 2. Cesium–137 concentrations in pasteurized milk, July 1965

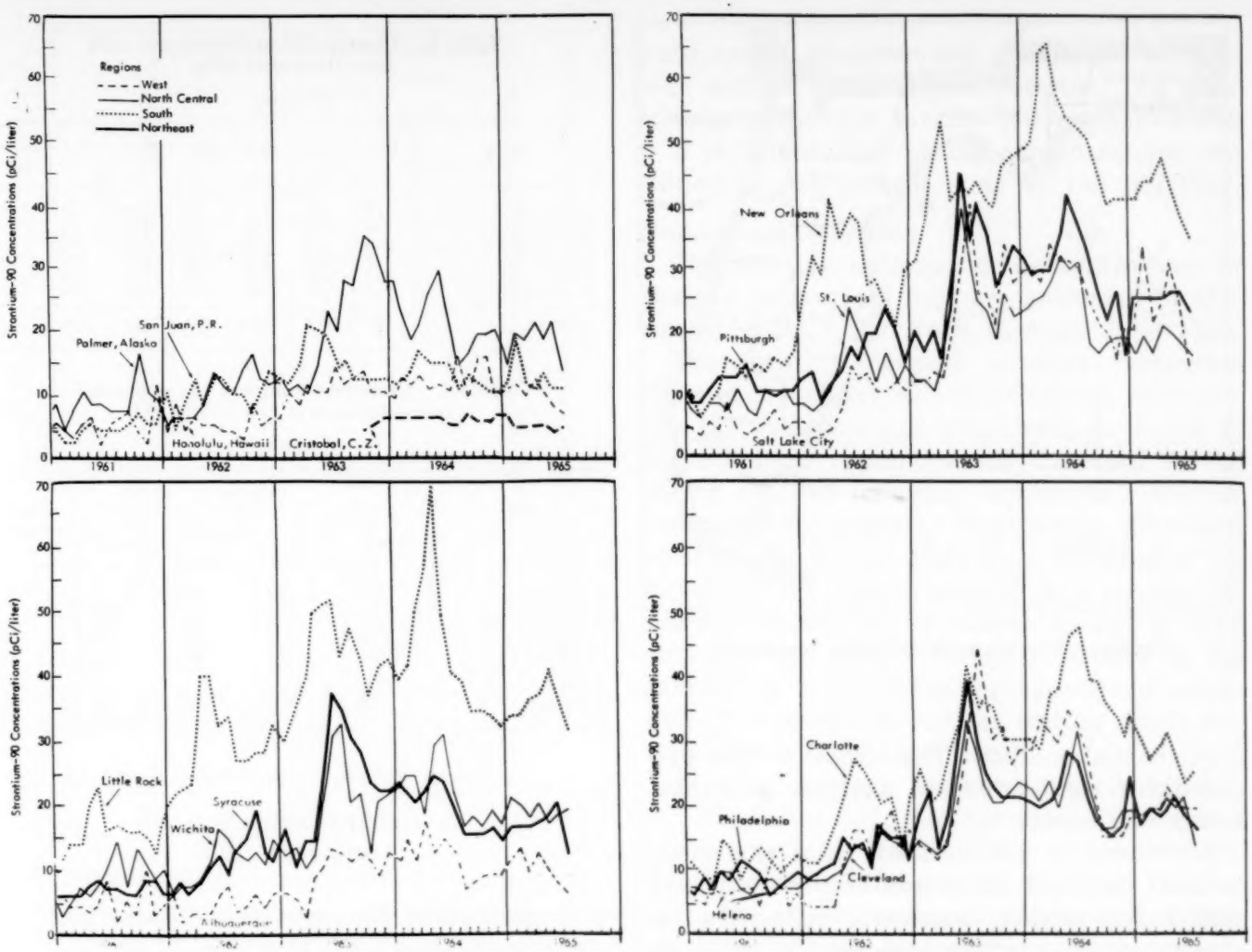

Figure 3. Strontium–90 concentrations in pasteurized milk, 1961—July 1965

2. Florida Milk Network July–December 1964 and Annual Summary

Division of Radiological and Occupational Health, Florida State Board of Health

The Florida State Board of Health began sampling raw milk for iodine–131 analysis in two major areas of the State in November 1962. The program has since been expanded to include the analysis of milk for strontium–89, strontium–90, and cesium–137, in addition to iodine–131. Raw milk samples are received from the six areas shown in figure 4. Samples for iodine–131 analysis are taken from a tank truck, the route of which passes by farms widely dispersed over the area represented. Where there is no route representative of a large portion of an area, samples collected from selected farms are combined. When iodine–131 is detectable in milk, samples are collected weekly. In the interest of maintaining an active standby capability, samples are now collected and analyzed for this nuclide on a monthly basis. A regional State Board of Health Laboratory is located in each of the six areas referred to in this report. Each laboratory prepares a monthly composite sample for its region by combining samples from 10 percent of the dairy farms selected at random. These composite samples are then sent to the State Radiological Health Laboratory in Orlando for analysis. Milk produced in the counties comprising each area is generally processed, marketed, and consumed in that area. These areas are characterized by differences in dairying practices related to the gradual transition from small farms in the West Florida region, where locally-grown feeds are used, to larger farms in the Southern areas, where different types of grass and predominantly purchased feeds are used.

Analytical procedures

Cesium–137 is determined by gamma scanning 1 liter of milk in a Marinelli beaker with a 4- x 4-inch sodium iodide crystal and

Figure 4. Florida milk sampling locations

multichannel analyzer. The sample is then prepared for radiostrontium analysis by evaporating and ashing.

Strontium is precipitated as a carbonate, counted for total radiostrontium activity, and stored for yttrium ingrowth. After the ingrowth period, yttrium is separated with carrier as an oxalate and counted in a low-background beta counter to determine strontium–90. Strontium–89 is then calculated as the difference between total radiostrontium and strontium–90 (as yttrium).

Results and discussion

During the period July to December 1964, strontium–89 and iodine–131 concentrations remained below the detectable level of 5 pCi/liter. Strontium–90 analyses were not performed routinely during the last half of 1964.

Table 5 presents the monthly average cesium–137 concentrations in Florida raw milk from July through December 1964.

A summary of 1964 observed cesium–137 concentrations is presented in table 6 as quarterly and yearly averages for each area and for the State. Following the mainland China nuclear test of October 16, 1964, raw milk samples were collected every other day at Pensacola from October 23 to November 12, 1964.

Table 5. Cesium-137 in Florida raw milk July–December 1964

(pCi/liter)

Area	July	Aug	Sept	Oct	Nov	Dec	Average
West Fla	169	174	145	169	106	77	140
North Fla	214	162	156	146	134	91	150
Northeast Fla	335	322	240	209	199	120	238
Central Fla	346	342	229	356	347	188	303
Tampa Bay	283	238	273	266	198	168	238
Southeast Fla	284	296	266	231	211	179	244
Average	272	256	218	231	199	137	219

Table 6. Quarterly summary of cesium-137 concentrations in raw milk, 1964

(pCi/liter)

Area	Quarter				Annual average
	First	Second	Third	Fourth	
West Fla	205	260	163	117	186
North Fla	183	267	177	124	188
Northeast Fla	179	249	299	176	226
Central Fla	255	335	306	300	299
Tampa Bay	203	257	265	211	234
Southeast Fla	212	254	278	207	238
Average	206	270	248	189	228

3. Indiana Milk Network April–June 1965

Bureau of Environmental Sanitation
Indiana State Board of Health

The Indiana State Board of Health began sampling pasteurized milk for radionuclide analysis in September 1961. To analyze radionuclide concentrations in Indiana milk effectively, the State was geographically divided into five major milksheds; northeast, northwest, central, southeast, and southwest (figure 5). One large dairy within each milkshed was assumed to be representative for sampling purposes.

The milk samples are routinely analyzed for strontium–89, strontium–90, cesium–137, iodine–131, and barium–140. Initially, cesium–137, iodine–131, and barium–140 were analyzed weekly. When iodine–131 concentrations exceeded 100 pCi/liter, the sampling frequency was increased. Since August 1963, because of the continued low concentrations of short-lived radionuclides, the sampling frequency was reduced to once a month for the northeast, southeast, and southwest milksheds. Monthly analyses for strontium–89 and strontium–90 are performed for each station.

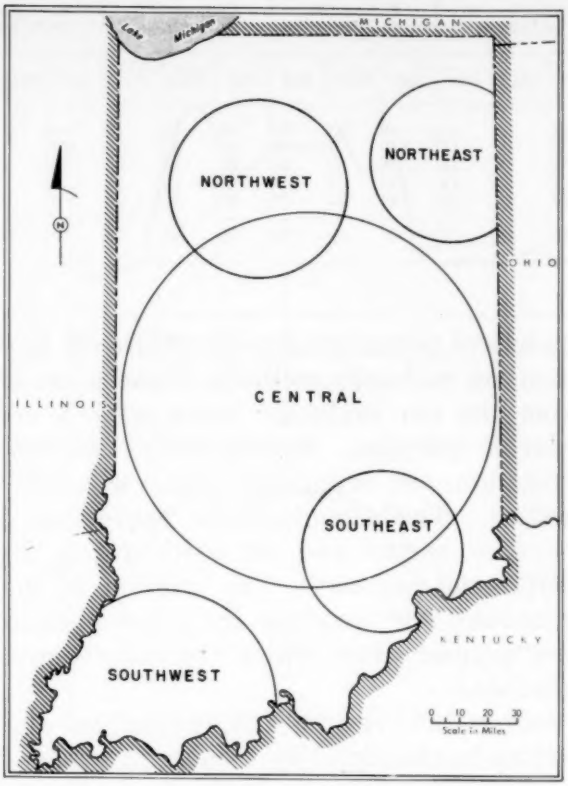

Figure 5. Indiana milk sampling locations

Analytical procedures

Strontium–89 and strontium–90 concentrations in milk samples are determined by ion exchange separation (*2*, *3*) and subsequent low-background beta counting. The limits of detectability for strontium–89 and strontium–90 are 5 and 1 pCi/liter, respectively. Cesium–137, iodine–131, and barium–140 are determined by gamma scintillation spectrometry. The counting system consists of a 512–channel pulse-height analyzer and a shielded 4- x 4-inch sodium iodide crystal detector. The limit of detectability for barium–140 and iodine–131 is 5 pCi/liter. Cesium–137 concentrations are subject to a 6 percent error at 100 pCi/liter.

Results and discussion

The monthly radionuclide concentrations in Indiana pasteurized milk are presented by station in table 7 for April through June 1965.

The monthly network average concentrations of strontium–89, strontium–90, and cesium–137 are presented graphically in figure 6. Strontium–89 concentrations increased in the spring of 1962 following atmospheric nuclear testing. Subsequently, these levels decreased until by the end of 1963 they were below detectable levels. Strontium–90 and cesium–137 concentrations also increased following 1962 atmospheric nuclear testing to a peak in the summer of 1963. With the subsequent cessation of atmospheric nuclear testing, these levels began a downward trend.

Monthly average concentrations in strontium–89, iodine–131, and barium–140 increased above detectable levels in May and June 1965. This increase is presumed to have resulted from the second mainland China test of May 14, 1965, and can be expected to decrease subsequently.

During the period covered, concentrations did not reach levels requiring countermeasures based upon health implications. Criteria for such remedial action have been outlined by the Federal Radiation Council (*4*).

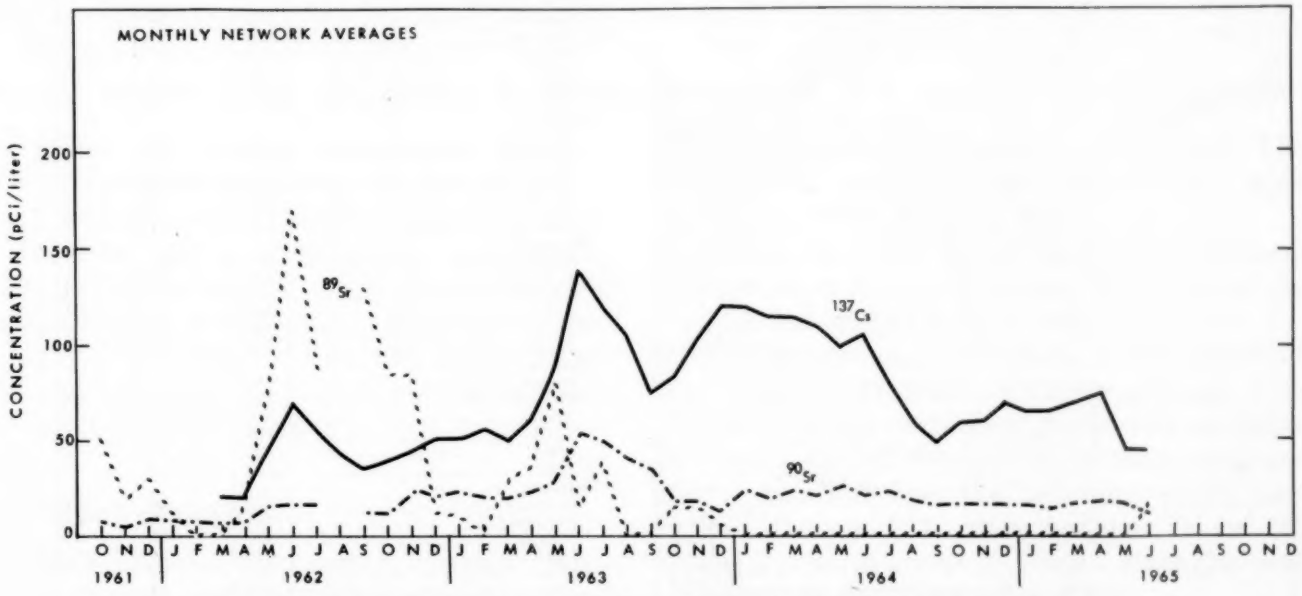
MONTHLY NETWORK AVERAGES
CONCENTRATION (pCi/liter)
200
150
100
50
0
89Sr
137Cs
90Sr
O N D J F M A M J J A S O N D J F M A M J J A S O N D J F M A M J J A S O N D J F M A M J J A S O N D
1961
1962
1963
1964
1965

Figure 6. Radionuclide concentrations in Indiana Pasteurized Milk Network, 1961—June 1965

Table 7. Radionuclide concentrations in Indiana milk, April–June 1965

Sampling location	Calcium (g/liter)			Potassium-40 (pCi/liter)			Strontium-89 (pCi/liter)			Strontium-90 (pCi/liter)			Iodine-131 (pCi/liter)			Cesium-137 (pCi/liter)			Barium-140 (pCi/liter)		
	Apr	May	Jun	Apr	May	Jun	Apr	May	Jun	Apr	May	Jun	Apr	May	Jun	Apr	May	Jun	Apr	May	Jun
Northeast	1.05	1.08	1.13	1,230	1,410	1,340	0	0	10	14	13	11	0	80	20	85	40	45	0	20	0
Southeast	NS	1.12	1.17	NS	1,200	1,350	NS	0	10	NS	16	11	NS	0	0	NS	45	35	NS	0	0
Central	1.11	1.10	1.13	1,310	1,330	1,380	0	0	20	14	13	11	0	10	30	65	45	50	0	0	10
Southwest	1.12	1.12	1.15	1,300	1,340	1,350	0	0	15	18	15	14	0	0	0	45	40	35	0	0	0
Northwest	1.11	1.08	1.13	1,330	1,330	1,420	0	0	20	14	15	10	0	20	40	80	55	55	0	10	10
Average	1.10	1.10	1.14	1,290	1,320	1,370	0	0	15	15	14	11	0	20	20	70	45	45	0	10	0

NS, no sample taken.

4. Michigan Milk Network January–June 1965

Division of Occupational Health
Michigan Department of Health

The Michigan Department of Health began sampling pasteurized milk for radionuclide analyses in November 1962. Under this program, weekly pasteurized milk samples are collected in the seven major milk producing areas in the State: Charlevoix, Detroit, Grand Rapids, Lansing, Marquette, Monroe, and Saginaw (figure 7). Daily samples within each area are composited in proportion to sales volume.

Analytical procedures

The resultant samples are analyzed weekly for strontium–90, iodine–131, cesium–137, potassium–40, and barium-lanthanum–140. The analytical procedure for strontium–90 is based upon ion exchange methods. Cations are eluted from the ion exchange resin with a sodium chloride solution. Subsequently, calcium and strontium are separated using a nitric acid method. The strontium is scavenged from the rare earths and set aside for a 2-week yttrium–90 ingrowth. The yttrium–90 is then separated and counted in a low-background beta counter after which the strontium–90 is calculated.

Iodine–131, cesium–137, potassium–40, and barium-lanthanum–140 concentrations are

Figure 7. Michigan milk sampling stations

Table 8. Radionuclides in seven Michigan milk producing areas, January–June 1965 [a]

(pCi/liter)

Station and month	Potassium-40	Strontium-90	Iodine-131	Cesium-137
Charlevoix:				
January	1,310	25	<14	95
February	1,370	30	<14	94
March	1,330	28	<14	95
April	1,330	25	<14	87
May	1,380	26	<14	87
June	1,310	21	19	61
Detroit:				
January	1,350	18	<14	69
February	1,370	19	<14	70
March	1,320	21	<14	69
April	1,310	16	<14	69
May	1,330	18	<14	61
June	1,300	17	21	38
Grand Rapids:				
January	1,370	20	<14	71
February	1,380	21	<14	72
March	1,330	21	<14	78
April	1,330	19	<14	74
May	1,360	21	<14	71
June	1,290	20	18	53
Lansing:				
January	1,450	18	<14	69
February	1,340	18	<14	65
March	1,360	20	<14	74
April	1,350	18	<14	70
May	1,330	19	<14	58
June	1,340	17	21	43
Marquette:				
January	1,380	NA	<14	144
February	1,360	42	<14	145
March	1,350	44	<14	148
April	1,360	34	<14	152
May	1,430	38	<14	140
June	1,350	36	<14	106
Monroe:				
January	1,320	12	<14	48
February	1,320	14	<14	54
March	1,300	14	<14	55
April	1,320	12	<14	53
May	1,350	14	<14	50
June	1,340	16	18	37
Saginaw:				
January	1,350	16	<14	66
February	1,360	16	<14	67
March	1,360	19	<14	65
April	1,330	14	<14	69
May	1,330	17	<14	67
June	1,320	14	<14	48

NA, no analyses performed
[a] Barium-lanthanum-140 values all less than 4 pCi/liter.

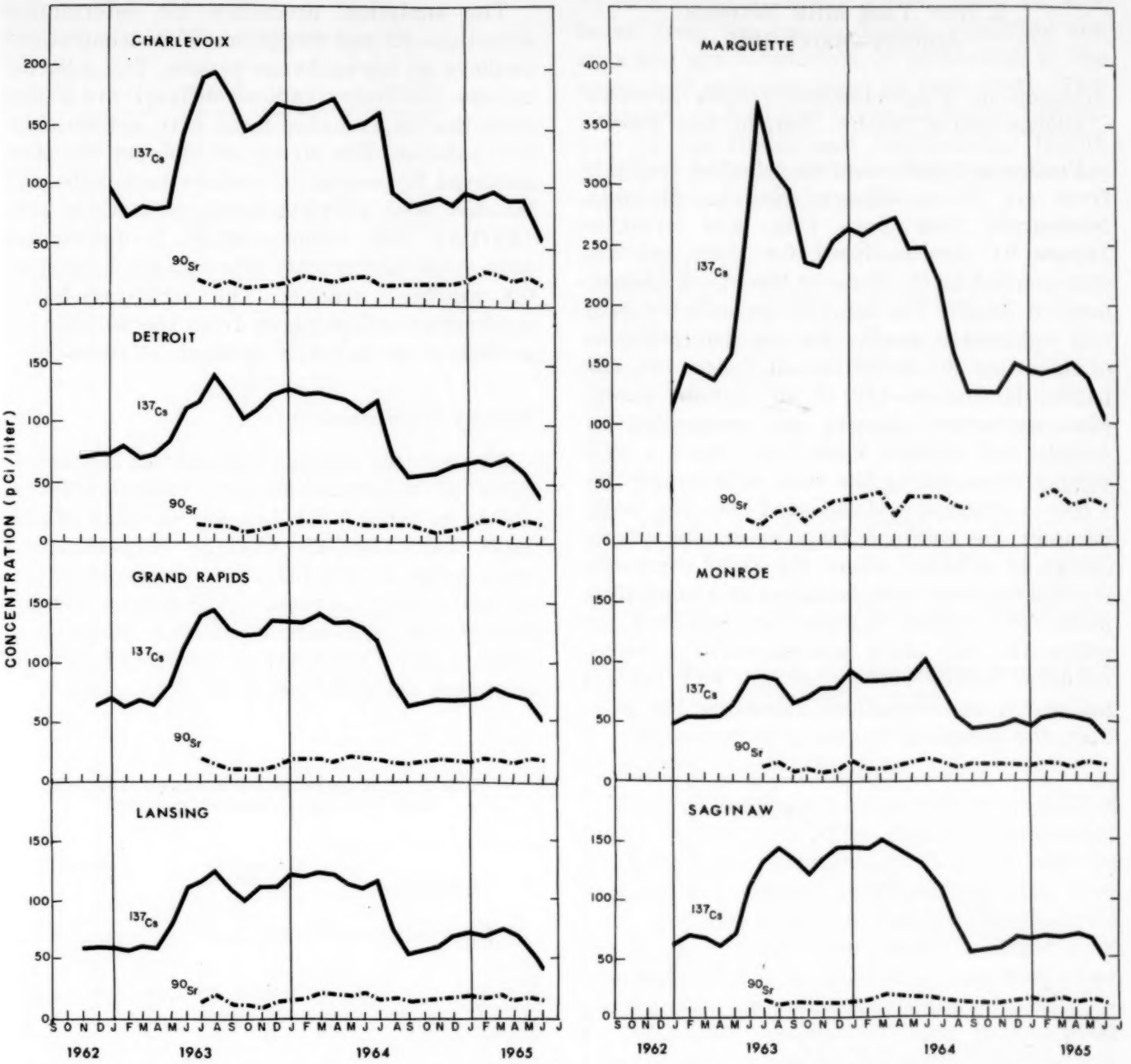

Figure 8. Monthly average concentrations of strontium–90 and cesium–137 in Michigan milk, 1962—June 1965

determined by gamma-scintillation spectrometry. The counting system consists of a 4- x 4-inch sodium iodide (thallium activated) crystal detector and a 400-channel pulse-height analyzer. The minimum levels of detectability are as follows: strontium–90, 9 pCi/liter; cesium–137, 5 pCi/liter; iodine–131, 14 pCi/liter; potassium–40, 80 pCi/liter; and barium-lanthanum–140, 4 pCi/liter.

Results and discussion

Table 8 presents the monthly average radionuclide concentrations in Michigan pasteurized milk for the period January to June 1965. The results are given for the seven major milk producing areas in the State.

During this period, iodine–131 and barium-lanthanum–140 concentrations in general remained below detectable levels.

To visualize general trends in strontium–90 and cesium–137 levels, monthly average concentrations are presented graphically in figure 8. Strontium–90 concentrations have remained fairly constant since the beginning of the program. Cesium–137 concentrations have followed a general decreasing trend since the peak observed in the summer of 1963.

5. New York Milk Network January–March 1965

Division of Environmental Health Services
Department of Health, State of New York

Pasteurized milk samples collected routinely from six cities—Albany, Buffalo, Massena, Newburgh, New York City, and Syracuse (figure 9)—are analyzed for their radionuclide content by the State of New York Department of Health. The samples are collected daily and composited weekly for the determination of strontium–89, strontium–90, iodine–131, and barium-lanthanum–140 at all stations except Massena, where samples are composited biweekly, and at New York City, where a milk sample representing the total milk supply for 1 day is obtained and analyzed once per week. Samples are obtained from processing plants except at Albany, where the daily composite is prepared from samples taken at a marketing point. The Albany samples are analyzed for iodine–131 and other gamma-emitting radionuclides. In the event that any city reports iodine–131 concentrations exceeding 100 pCi/liter, the sampling frequency is increased.

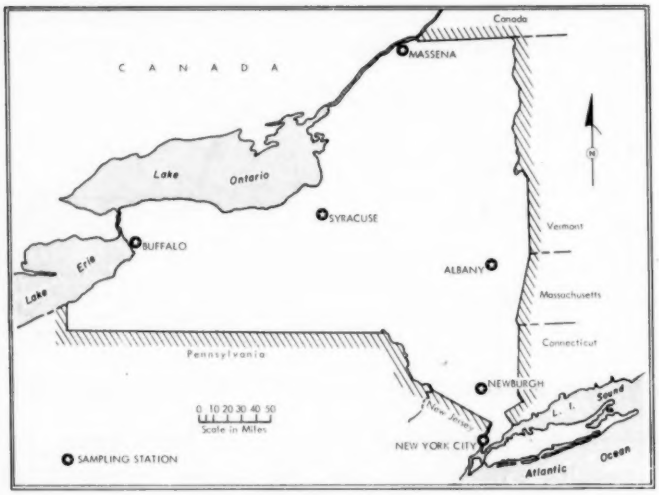

Figure 9. New York sampling stations

Analytical procedures

Gamma-emitting radionuclide concentrations in milk are determined by scintillation spectrometry and the application of a matrix method of analysis (*5*) to the resultant spectral data.

The analytical procedure for determining strontium–89 and strontium–90 concentrations employs an ion exchange system. The collected cations (including radiostrontium) are eluted from the ion exchange resin with sodium chloride solution. The strontium isotopes are then gathered by means of sodium carbonate and isolated with ethylenediaminetetraacetic acid (EDTA). The radiostrontium is determined with a low-background beta counter having an 0.8 mg/cm^2 window. The strontium–90 is determined differentially from the 40-hour ingrowth of its daughter product, yttrium–90.

Results and discussion

The monthly average radionuclide concentrations of strontium–90 and cesium–137 are shown in table 9 for January through March 1965. Strontium–89 average concentrations were below 3 pCi/liter. Strontium–90 values showed a slight increase in this quarter. During this period, the iodine–131 and barium-lanthanum–140 concentrations remained below the minimum detectable level of 20 pCi/liter.

Table 9. Average radionuclide concentrations in New York milk, January–March 1965

Sampling station	Strontium-90 (pCi/liter)			Cesium-137 (pCi/liter)		
	Jan	Feb	Mar	Jan	Feb	Mar
Albany	13	14	17	72	63	51
Buffalo	13	10	13	67	68	76
Massena	13	20	NS	92	102	104
Newburgh	17	18	19	79	65	NS
New York City	16	17	19	74	69	66
Syracuse	9	15	NS	56	63	60
Average	13.5	15.7	17.0	74.0	71.7	71.4

NS indicates no sample collected.

The cesium–137 and iodine–131 concentrations since September 1961 are represented in figure 10. Despite the slight increase shown in this report, the general long-range trend in cesium–137 concentrations since the atmospheric test ban treaty is one of decreasing levels. Seasonal variations have been apparent during this period.

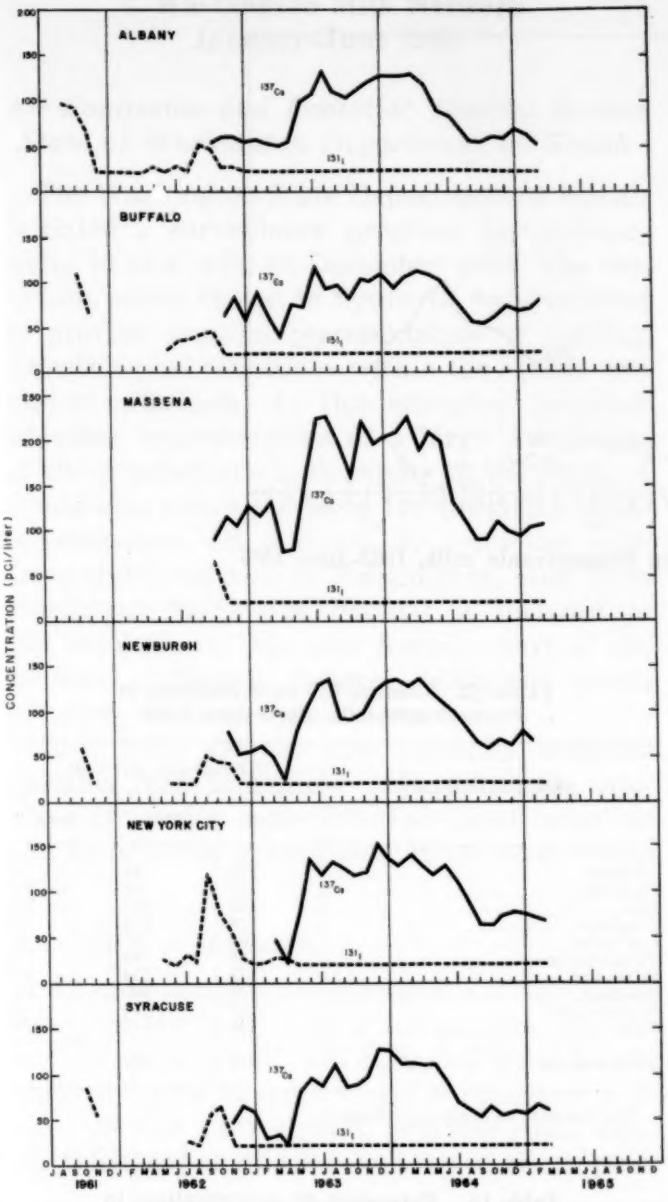

Figure 10. Cesium–137 and iodine–131 in New York milk, September 1961–March 1965

6. Pennsylvania Milk Network April–June 1965

Bureau of Environmental Health
Pennsylvania Department of Health

Samples of pasteurized milk are routinely collected from 10 major milk consumption areas throughout Pennsylvania (figure 11). Two samples per week are collected in Philadelphia and Pittsburgh, while weekly composite samples are collected from the other eight stations. At each sampling location subsamples are collected from the major dairies supplying the area and are composited in proportion to the amount of milk processed by each dairy. This composite is then sent to the Radiation Laboratory of the Division of Occupational Health in Harrisburg where the weekly samples are combined for monthly analysis.

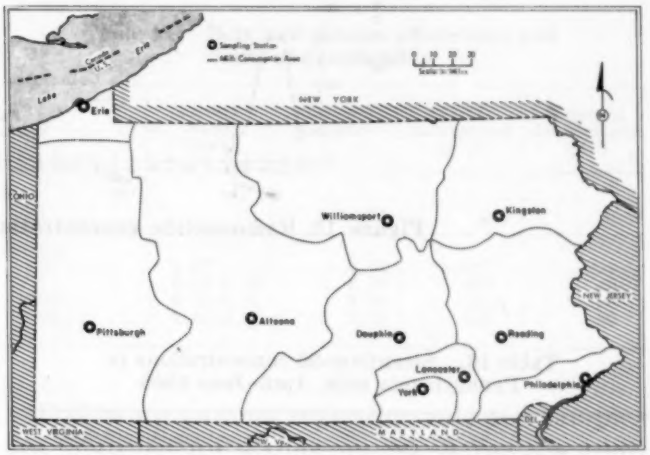

Figure 11. Pennsylvania milk sampling stations and milk consumption areas

Analytical procedures

Strontium–90 analyses have been carried out since April 1963. The chemical separation technique for strontium–90 is essentially the ion exchange method described by Porter, *et al.* (*2*). One liter of milk is passed through an ion exchange column; yttrium–90 is eluted from the resin and is counted in an automatic low-background proportional counter.

Iodine–131, cesium–137, and potassium–40 concentrations are determined by gamma scintillation spectrometry. The resultant gamma spectra are evaluated, using a matrix system of analysis.

Results

The average monthly strontium–90 concentrations in Pennsylvania pasteurized milk for April to June 1965 are shown in table 10, and presented graphically in figure 12.

Iodine–131 was observed in May and June 1965 as shown in table 11 and plotted in figure 12. Cesium–137 and potassium–40 concentrations are given in tables 12 and 13, respectively.

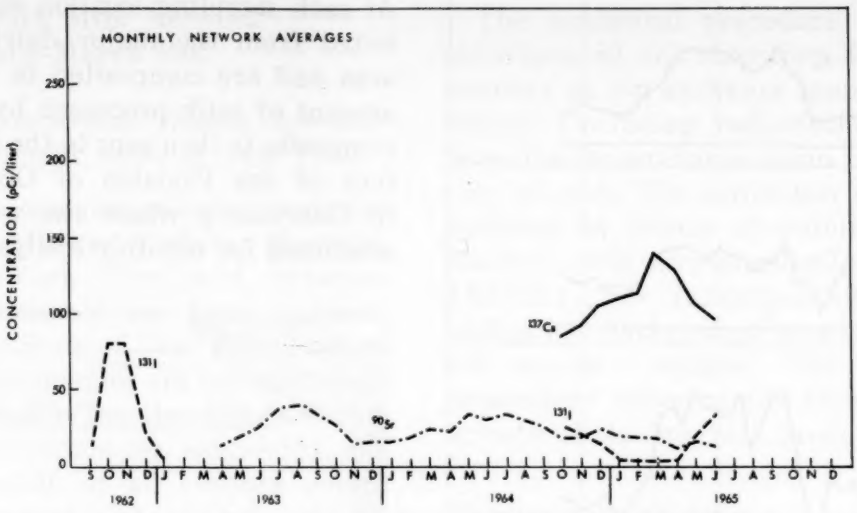

Figure 12. Radionuclide concentrations in Pennsylvania milk, 1962–June 1965

Table 10. Strontium-90 concentrations in Pennsylvania milk, April–June 1965

Sampling location	Strontium-90, pCi/liter		
	April	May	June
Altoona	17	16	13
Dauphin	15	18	14
Erie	17	20	19
Kingston	17	16	19
Lancaster	12	20	12
Philadelphia	16	19	15
Pittsburgh	27	23	23
Reading	17	20	13
Williamsport	13	17	13
York	19	19	12
State average	17	19	15

Table 11. Iodine-131 concentrations in Pennsylvania milk, April–June 1965

Sampling location	Iodine-131, pCi/liter		
	April	May	June
Altoona	<10	16	22
Dauphin	<10	40	24
Erie	<10	17	NA
Kingston	<10	34	30
Lancaster	<10	38	<10
Philadelphia	<10	<10	66
Pittsburgh	<10	10	45
Reading	<10	17	37
Williamsport	<10	14	40
York	<10	32	16
State average	<10	22	31

NA indicates no analysis performed.

Table 12. Cesium-137 concentrations in Pennsylvania milk, April–June 1965

Sampling location	Cesium-137, pCi/liter		
	April	May	June
Altoona	120	98	80
Dauphin	122	104	90
Erie	149	102	NA
Kingston	138	136	104
Lancaster	124	97	90
Philadelphia	136	99	106
Pittsburgh	133	134	108
Reading	120	100	105
Williamsport	118	94	89
York	130	110	79
State average	129	107	95

NA indicates no analysis performed.

Table 13. Potassium-40 concentrations in Pennsylvania milk, April–June 1965

Sampling location	Potassium-40, pCi/liter		
	April	May	June
Altoona	905	1,002	939
Dauphin	1,065	1,098	1,096
Erie	1,130	1,020	NA
Kingston	1,125	1,109	1,064
Lancaster	1,141	1,091	1,005
Philadelphia	1,680	1,083	1,111
Pittsburgh	1,110	1,113	1,059
Reading	1,113	1,004	1,124
Williamsport	945	1,002	1,004
York	1,020	1,154	1,039
State average	1,123	1,068	1,049

NA indicates no analysis performed.

Previous coverage in *Radiological Health Data:*

Period	*Issue*
September 1962–November 1963	*March 1964*
December 1963–March 1964	*July 1964*
April–June 1964	*October 1964*
August–September 1964	*February 1965*
October–December 1964	*June 1965*
January–March 1965	*September 1965*

7. Washington Milk Network January–June 1965

Air Sanitation and Radiation Control Section
State of Washington Department of Health

The Washington State Department of Health initiated a surveillance program for radioactivity in raw milk in December 1962. The collection points shown in figure 13 were selected to provide samples representative of varying climatological conditions within the State's two major milksheds. As this sampling program provides representation of a large percentage of the population's milk supply in the State, it would also provide a basis for initiating counter-measures. In addition to sampling milk from eight locations in Washington, milk from Northwest Idaho (Sandpoint) is included in the network, as this area forms a part of the Spokane milkshed. Several additional points are sampled from time to time.

Raw milk samples are normally collected monthly from individual tankers, but the sampling frequency is flexible. Increased sampling can be initiated immediately if the need should arise.

Analytical procedures

All milk samples are placed in a 2-liter stainless steel Marinelli beaker and gamma scanned for 100 minutes with a 512-channel pulse-height analyzer. This beaker, which is placed on a 3- x 3-inch NaI (Tl) crystal, provides equal sample thickness on all sides and top of the crystal. A 4 x 4 matrix is used to analyze the spectra and provide rapid quantitative results for iodine–131, barium-140, cesium–137, and potassium–40.

As a check of analyzer stability, daily 1-minute counts of cesium–137 are made with the same source. Gamma efficiencies and detectability limits are given in table 14 for this system.

Table 14. Beta and gamma efficiencies and limits of detectability

	Energy band (MeV)	Efficiency (percent)	Average background (pCi)	Limits of detectability (pCi)
Beta				
^{90}Y		37	0.6	0.24
Gamma				
^{40}K	1.37–1.55	0.18	2,277	325
^{131}I	0.33–0.40	4.20	300	20
^{137}Cs	0.62–0.72	2.60	246	30
^{140}Ba	0.46–0.57	3.50	326	30

Following gamma scanning, selected samples are analyzed for strontium–90. After the addition of strontium carrier, the milk proteins are precipitated with trichloroacetic acid. After filtration, oxalic acid is added to precipitate the alkaline earths as oxalates (pH 3.0). The oxalates are ashed, dissolved in 6*N* hydrochloric acid, and the pH is adjusted to 1.4. A double extraction is performed, using equal portions of 20 percent (by volume) di-(2-ethylhexyl) phosphoric acid (HDEHP) in toluene. This effectively removes rare-earth activity, including yttrium–90, leaving the strontium–90 in the

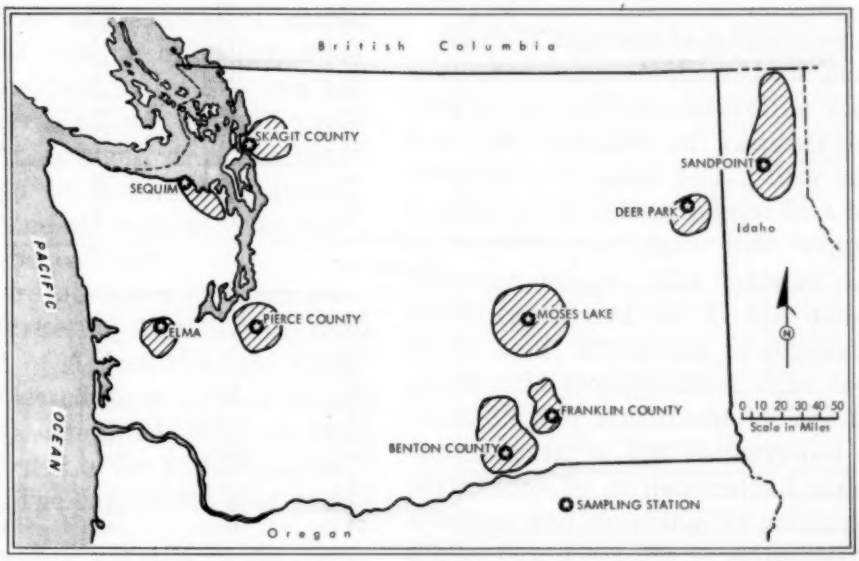

Figure 13. Washington milk sampling locations

sample. This sample is stored a minimum of 2 weeks to allow yttrium–90 to ingrow. Subsequently, the yttrium–90 is extracted with 5 percent (by volume) HDEHP in toluene. The organic layer is scrubbed with a HCl solution (pH 1.4) and back extracted with 3*N* HNO_3. The resulting nitric acid solution is evaporated and yttrium–90 counted. From this, the strontium–90 activity is calculated. The yttrium–90 counting is done in a low-background, gas-flow, internal proportional counter. The efficiency and detectability limit of the counter for yttrium–90 are given in table 14.

As a part of the laboratory's quality control program, some samples are split and analyzed several times. Split samples are also exchanged with State, Federal, and university laboratories for analysis and comparisons.

Results

Table 15 presents the monthly average radionuclide concentrations in Washington raw milk for January through June 1965. The results are given for representative production areas within the two major milksheds.

During this period fresh fission product concentrations remained below detectable levels, except in three samples in May and June, as shown in table 16. These samples are presumed to reflect the second mainland China atmospheric nuclear detonation of May 14, 1965.

Zinc–65 was observed in Benton and Franklin Counties on four occasions during this period (table 17). This nuclide has shown up routinely in these counties and results from irrigation of some pasture land with Columbia River

Table 15. Radionuclide concentrations, Washington milk, January–June 1965

(pCi/liter)

Potassium-40

Sampling location	January	February	March	April	May	June
Benton County	NS	1,230	NS	1,340	1,130	NS
Deer Park	NS	1,220	1,280	1,230(3)	1,180(2)	1,160(2)
Elma	1,100	1,220	1,200	1,110	1,260(2)	1,170(2)
Franklin County	1,240	NS	1,260	1,110(2)	1,230	1,190(2)
Moses Lake	NS	1,230	1,240	1,210(3)	1,180(2)	1,150(2)
Pierce County	1,220	1,230	1,330	1,190	1,230(2)	1,130(2)
Sandpoint	NS	1,260	1,180	1,160(3)	1,270(2)	1,290(2)
Sequim	1,210	1,060	1,180	1,170	1,190(2)	1,170(2)
Skagit County	1,120	1,110	1,100	1,180	1,150(2)	1,130(2)
Average	1,180	1,200	1,220	1,190	1,200	1,170
Maximum	1,240	1,260	1,330	1,340	1,310	1,320
Minimum	1,100	1,060	1,100	1,060	1,110	1,030

Strontium-90

Benton County	NS	10	NS	10	NA	NS
Deer Park	NS	24	24	20	22	22
Elma	23	21	17	12	37	29
Franklin County	10	NS	10	NA	9	7
Moses Lake	NS	10	9	10	10	7(2)
Pierce County	15	11	14	13	29	18
Sandpoint	NS	27	28	29	38(2)	31(2)
Sequim	19	20	24	21	19	11
Skagit County	22	21	22	19	25	26
Average	18	18	18	17	24	19
Maximum	23	27	28	29	38	32
Minimum	10	10	9	10	9	7

Cesium-137

Benton County	NS	47	NS	59	27	NS
Deer Park	NS	148	137	135(3)	116(2)	96(2)
Elma	135	152	82	51	143(2)	113(2)
Franklin County	54	NS	43	55(2)	42	33(2)
Moses Lake	NS	61	50	50(3)	50(2)	32(2)
Pierce County	129	71	93	103	102(2)	98(2)
Sandpoint	NS	142	131	153(3)	152(2)	144(2)
Sequim	135	114	108	134	77(2)	51(2)
Skagit	124	135	102	101	101(2)	83(2)
Average	115	109	93	93	90	81
Maximum	135	152	137	164	159	145
Minimum	54	47	43	47	27	26

Key to symbols: ( ), number of samples in average.
NS, no sample collected.
NA, no analysis performed.

Table 16. Milk samples containing iodine-131

Sampling station	Date (1965)	Iodine-131 (pCi/liter)
Sandpoint	May 25	19
Moses Lake	June 1	16
Sandpoint	June 2	19

Table 17. Milk samples containing zinc-65

Sampling station	Date (1965)	Zinc-65 (pCi/liter)
Franklin County	January 6	110
Benton County	February 9	57
Franklin County	March 3	53

water which has been shown to contain this contaminant.

To visualize general trends in strontium–90 and cesium–137 concentrations in Washington milk, monthly average network concentrations are presented graphically in figure 14. These averages are highly artificial, as they are not weighted according to the volume of milk from each production area, nor do they take into consideration the absence of samples from several stations for a particular month.

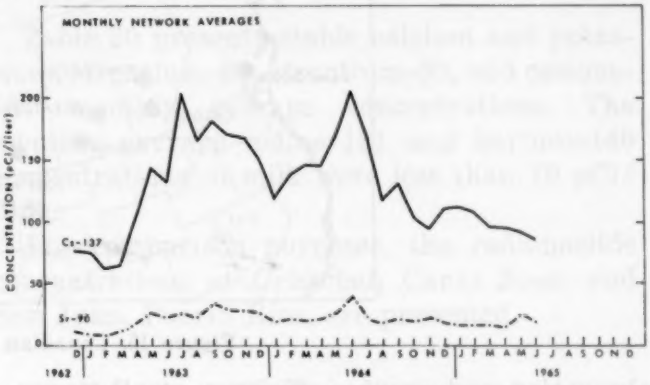

Figure 14. Radionuclides in Washington milk, 1962–June 1965

8. Canadian Milk Network[3] July 1965

Radiation Protection Division, Department of National Health and Welfare, Ottawa, Canada

The Radiation Protection Division of the Department of National Health and Welfare began monitoring milk for strontium–90 in November 1955. At first, analyses were carried out on samples of powdered milk obtained from processing plants. However, since January 1963 liquid whole milk has been analyzed instead. With this change, more representative samples of milk consumed can be obtained, and milk sampling locations (see figure 15) can be chosen in the same areas as the air and precipitation stations. At present, the analyses include determinations of iodine–131, strontium–89, cesium–137, and strontium–90 as well as stable potassium and calcium.

The milk samples are obtained through the cooperation of the Marketing Division of the Canadian Department of Agriculture. At each station samples are collected three times a week from selected dairies, combined into weekly composites, and forwarded to the radiochemical laboratory in Ottawa. The contribution of each dairy to the composite sample is directly proportional to its volume of sales. In most cases, a complete sample represents over 80 percent of the milk processed and distributed in the area. Several of the weekly samples are randomly selected and analyzed for iodine–131. The results of the spot checks for iodine–131 will not be reported unless there is evidence that the levels are rising. A monthly composite of the samples is analyzed for strontium–90, cesium–137, and stable potassium and calcium.

Analytical methods

Since it represents a more sensitive method, radiochemistry, rather than gamma spectrometry, is used to determine iodine–131 (*6*). For the analysis of radiostrontium, carrier strontium is added to a 1-liter sample of milk, and the milk is then placed in a tray lined with a polyethylene sheet and evaporated under infrared lamps. The residue is ashed in a muffle furnace at 450°C. and dissolved in dilute nitric acid; strontium is separated by fuming nitric acid precipitation. The combined strontium–89 and strontium–90 are determined by counting in a low-background beta counter. Strontium–90 is determined separately by extracting and counting its yttrium–90 daughter, while strontium–89 is estimated by difference from the total radiostrontium measurement. Appropriate corrections are made for self-

[3] Prepared from information and data in the June 1965 monthly report "Data from Radiation Protection Programs," Canadian Department of National Health and Welfare, Ottawa, Canada.

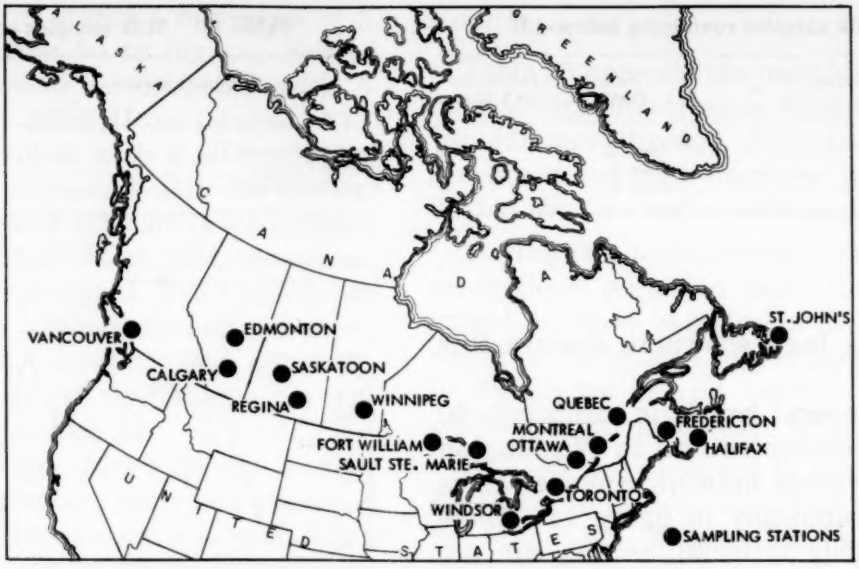

Figure 15. Canadian milk sampling stations

absorption and counter efficiency at all stages. Calcium is determined by flame photometry.

Cesium–137 is determined by gamma spectrometry using a scintillation crystal and a multichannel pulse-height analyzer. A sample consisting of 4.5 liters of milk is placed in a sample tray constructed in the form of an inverted well to accommodate the 5- by 4-inch sodium iodide crystal detector. The sample is counted for 100 minutes and the gamma spectrum is then recorded. Estimates are made of the potassium–40 and cesium–137 content of the milk by comparison of the spectrum with the spectra of standard preparations of these two radionuclides. With this method the potassium–40 concentration is determined and the Compton contribution of this radionuclide to the cesium–137 photopeak is subtracted to obtain the cesium–137 concentration. The stable potassium content is estimated from the potassium–40 concentration.

Sources of error

In the iodine and strontium determinations, tests indicate that the statistical error (95 percent confidence level) in the chemical operations involved is about plus-or-minus 10 percent. This value is independent of the concentration of the radionuclide in the milk because it depends only on the recovery of the carrier. In the determination of cesium–137 this factor is not involved.

The chemical procedures error must be combined with the counting error which depends primarily on the concentration of the nuclide in the sample, the background radiation, and the length of time the sample and background are counted. This counting error has been evaluated mathematically for the particular counting arrangement used.

The overall errors, estimated on the basis indicated above, are given in table 18.

Table 18. Total error for various radionuclide concentrations in milk [a]

Nuclide	Error for 10 pCi/liter	Error for 50 pCi/liter	Error for 100 pCi/liter
Strontium-89	±25%	±20%	±15%
Strontium-90	±15%	±10%	±10%
Iodine-131	±50%	±20%	±10%
Cesium-137	±60%	±20%	±10%

[a] All errors are 2σ values, representing 95 percent confidence levels.

Results

Table 19 presents monthly averages of strontium–90, cesium–137, and stable calcium and

Table 19. Stable elements and radionuclides in Canadian whole milk, July 1965 [a]

Station	Calcium (g/liter)	Potassium (g/liter)	Strontium-90 (pCi/liter)	Cesium-137 (pCi/liter)
Calgary	1.15	1.6	25.9	89
Edmonton	1.15	1.6	25.4	76
Ft. William	1.15	1.6	30.1	124
Fredericton	1.09	1.6	31.9	114
Halifax	1.18	1.6	31.4	102
Montreal	1.15	1.6	16.7	57
Ottawa	1.15	1.6	15.2	57
Quebec	1.16	1.6	31.8	94
Regina	1.00	1.6	23.1	58
St. John's, Nfld	1.23	1.6	53.4	204
Saskatoon	1.00	1.6	17.3	56
Sault Ste. Marie	1.16	1.7	33.2	126
Toronto	1.19	1.5	9.9	53
Vancouver	1.20	1.6	22.9	67
Windsor	1.25	1.6	9.4	38
Winnipeg	1.08	1.4	22.7	77
Average	1.14	1.6	25.0	87

[a] Due to insignificant levels of iodine-131 and strontium-89, the reporting of these radionuclides has been discontinued.

potassium in Canadian whole milk. Spot checks for iodine–131 and strontium–89 indicate that all samples had insignificant levels of these radionuclides.

The results show that radionuclide concentrations in Canadian whole milk remained well below the levels permissible on health grounds.

9. Pan American Milk Sampling Program July 1965

Pan American Health Organization, and Public Health Service

In accordance with a joint agreement, the Pan American Health Organization (PAHO) and the Public Health Service (PHS) developed a collaborative program for furnishing assistance to health authorities in the Americas engaged in developing programs in radiological health.

Under this agreement, the PHS Division of Radiological Health furnishes to PAHO, on a loan basis, limited quantities of essential items of equipment and the requisite laboratory services to establish a surveillance program.

Sampling procedure

Initially, air sampling stations were established in Chile, Jamaica, Peru, and Venezuela. In August 1963, the program was expanded to include a milk sampling station in Caracas, Venezuela. Between April 1964 and August 1964, additional stations were established in Jamaica at Kingston, Montego Bay, and Mandeville. Sampling varies according to local procedures.

Under the direction of the Venezuelan Institute for Scientific Investigation, weekly samples are collected, preserved with formaldehyde, and composited monthly.

Jamaica, under the direction of the Ministry of Health, collects one monthly composite on a rotating basis from one of the three principal milk areas: Montego Bay (Montpelier), Mandeville, and Kingston (Spanish Town). All samples are sent to the PHS Southeastern Radiological Health Laboratory for analyses.

Analytical procedures

Iodine–131 and cesium–137 are determined by gamma scintillation spectrometry. Strontium–89, strontium–90, and barium–140 are determined radiochemically (*3*). Analytical errors are discussed in the "Analytical Procedures, Pasteurized Milk Network," page 607.

Data presentation

Table 20 presents stable calcium and potassium, strontium–89, strontium–90, and cesium–137 monthly average concentrations. The monthly average iodine–131 and barium–140 concentrations in milk were less than 10 pCi/liter.

For comparison purposes, the radionuclide concentrations at Cristobal, Canal Zone, and San Juan, Puerto Rico, are presented.

Table 20. Stable element and radionuclide concentrations in milk, PAHO, July 1965

Sampling station	Calcium (g/liter)	Potassium (g/liter)	Strontium-89 (pCi/liter)	Strontium-90 (pCi/liter)	Cesium-137 (pCi/liter)
Canal Zone:					
Cristobal	1.10	1.5	<5	4	30
Jamaica:					
Kingston	NS	NS	NS	NS	NS
Mandeville	1.19	1.59	<5	9	190
Montego Bay	NS	NS	NS	NS	NS
Puerto Rico:					
San Juan	1.10	1.6	<5	10	45
Venezuela:					
Caracas	1.23	1.46	<5	3	15

NS indicates no sample collected during this period.

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(*4*) FEDERAL RADIATION COUNCIL. Background material for the development of radiation protection standards, Report No. 5. Superintendent of Documents, U.S. Government Printing Office, Washington, D.C. 20402 (July 1964).

(*5*) KAHN, B., *et al.* Rapid methods for estimating fission product concentrations in milk. PHS Publication No. 999–R–2. Single copies available on request from Public Inquiries Branch, PHS, U.S. Department of Health, Education, and Welfare, Washington, D.C. 20201 (March 1963).

(*6*) DASGUPTA, A. K., and H. G. GREEN. A method for the radiochemical determination of iodine–131 in milk. RPD–23, Radiation Protection Division, Department of National Health and Welfare, Ottawa, Canada (October 1963).

APPLICATION OF RADIONUCLIDE CONCENTRATIONS IN MILK TO INTAKE GUIDES, JULY 1964–JULY 1965

Division of Radiological Health, Public Health Service

The concentrations of specific radionuclides in milk analyzed as part of the Pasteurized Milk Network (PMN) are reported on a monthly basis in *RHD*. In terms of radiological health surveillance activities, an important aspect of these data is the estimation of resultant radiation dose to population groups.

Approximate relationships between certain radionuclide intakes and dose have been applied to the formulation of daily intake guides (*1*) and permissible concentrations in selected environmental media (*2*). Although these guides are not themselves directly applicable to worldwide fallout, a comparison with environmental contamination levels does yield a measure of population dosage. In general, intake-dose and dose-biological effect relationships used in formulating the guides cited are based on continuous intake over an entire lifetime. However, for general surveillance purposes, yearly average intakes, used with discretion, may be compared directly with the levels adopted as lifetime intake guides. Thus, the radionuclide concentrations in milk, averaged over a year's time, together with milk consumption data, might be used in conjunction with the references cited above to approximate the radiation dose to a specific population group from a specific radionuclide. Table 1 presents annual averages of radionuclide concentrations in milk sampled by the PMN. Limited data are available for estimating the average daily milk consumption (on a volume basis) for specific age groups in the U.S. population (*3*, *4*).

Total dietary intake is of prime interest, and since the intake via milk consumption constitutes only a portion of the total radionuclide intake, the relationship of milk intake to total dietary intake is of importance in evaluating milk surveillance data. The Federal Radiation Council (*5*) notes: "A number of studies have shown that conservative estimates of the strontium–90 to calcium ratio in the total diet may be made by multiplying the ratio of strontium–90 to calcium in milk in a particular locality by 1.5."[1] Thus, a rough index of the total dietary intake of strontium–90 on an annual basis may be made from PMN annual averages by using this factor and the assumptions of approximately 1.2 grams of calcium per liter in PMN samples and a 1.0 gram daily intake of calcium.

In the case of iodine–131, milk can be considered the major source because of the rapid distribution and consumption of fresh milk. With most other foods, normal processing and distribution allows time for this short-lived nuclide to decay to insignificant levels.

The situation with respect to strontium–89 is more complicated. Its half-life of some 50 days makes it difficult to estimate the relative contribution made by sources other than milk to the total dietary intake.

The relative contribution of milk to the total dietary intake of cesium–137 is not well defined and depends principally on the amount of freshly deposited cesium–137 on products used for human and animal consumption, and the progress of cesium–137 through the food chain.

The data in table 1 are calculated as follows: results from all samples collected in each week (Sunday through Saturday) are averaged, and the averages for all weeks terminating in each of 12 consecutive months are averaged to obtain the annual average[2]. To obtain the annual average daily intake (pCi/day) of radionuclides from milk, the annual average concentration values (pCi/liter) in table 1 must be multiplied by the annual average daily consumption (liters/day) of milk.

Monthly variations of radionuclide concentrations in milk are influenced by a number of combined causes such as meteorologic conditions and dairying practices, apart from considerations of original sources of radionuclides. The moving yearly average (table 1) obtained by updating the previous 12-month average by 1 month, shows variations averaged over the year and tends to minimize purely seasonal variations.

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[1] This ratio may vary from 1 to 2, depending on changes in rate of fallout deposition and relative consumption of non-milk products whose contamination reflects temporal and local deposition patterns (*6*).

[2] Beginning with the October 1963 data, iodine–131 values of <10 pCi/liter are considered to be zero for averaging purposes; previously, 5 pCi/liter was used for calculating the averages.

Table 1. Average radionuclide concentrations in milk for the 12-month periods,[a] July 1964–June 1965 and August 1964–July 1965

Sampling locations		Strontium-89 (pCi/liter)		Strontium-90 (pCi/liter)		Iodine-131 (pCi/liter)		Cesium-137 (pCi/liter)	
		July '64–June '65	Aug. '64–July '65	July '64–June '65	Aug '64–July '65	July '64–June '65	Aug. '64–July '65	July '64–June '65	Aug '64–July '65
Ala:	Montgomery	3	3	19	19	0	0	56	53
Alaska:	Palmer	3	3	18	17	1	1	78	73
Ariz:	Phoenix	3	3	5	5	0	0	24	24
Ark:	Little Rock	4	5	36	35	2	2	70	68
Calif:	Sacramento	3	3	7	7	1	1	31	30
	San Francisco	3	3	8	8	0	0	32	31
C. Z:	Cristobal	3	3	5	5	0	0	44	42
Colo:	Denver	3	3	18	18	1	1	77	71
Conn:	Hartford	3	3	18	17	0	0	91	85
Del:	Wilmington	3	3	19	18	2	2	76	71
D. C:	Washington	3	3	17	17	2	2	56	53
Fla:	Tampa	3	3	14	13	0	0	200	194
Ga:	Atlanta	3	3	27	26	1	1	92	86
Hawaii:	Honolulu	3	3	11	11	0	0	66	64
Idaho:	Idaho Falls	3	3	21	20	2	2	100	93
Ill:	Chicago	3	3	17	16	2	2	81	75
Ind:	Indianapolis	3	3	17	16	4	4	65	60
Iowa:	Des Moines	4	4	22	22	6	6	60	56
Kans:	Wichita	3	3	18	18	2	2	48	46
Ky:	Louisville	4	4	24	23	2	2	50	47
La:	New Orleans	5	5	43	41	2	2	88	83
Maine:	Portland	3	3	24	23	2	2	144	137
Md:	Baltimore	3	3	19	18	2	2	61	58
Mass:	Boston	3	3	25	23	0	0	142	133
Mich:	Detroit	3	3	16	15	3	3	81	75
	Grand Rapids	3	3	19	18	2	2	90	84
Minn:	Minneapolis	6	7	26	26	6	5	92	86
Miss:	Jackson	3	3	34	33	1	1	64	60
Mo:	Kansas City	5	5	23	22	8	8	50	47
	St. Louis	3	4	18	18	4	4	49	45
Mont:	Helena	3	3	18	18	3	3	97	92
Nebr:	Omaha	3	3	21	20	4	4	60	56
Nev:	Las Vegas	3	3	7	7	2	2	45	42
N. H:	Manchester	3	3	25	25	1	1	164	154
N. J:	Trenton	3	3	16	16	1	1	77	72
N. Mex:	Albuquerque	2	2	10	10	1	1	42	39
N. Y:	Buffalo	3	3	17	16	0	0	100	93
	New York	3	3	21	20	2	2	102	95
	Syracuse	[b] 3	3	[b] 16	16	[b] 1	1	[b] 86	82
N. C:	Charlotte	3	3	32	30	1	1	74	69
N. Dak:	Minot	6	7	49	47	2	2	124	115
Ohio:	Cincinnati	3	3	18	17	2	2	59	55
	Cleveland	3	4	18	18	2	2	82	76
Okla:	Oklahoma City	4	4	19	19	2	2	49	46
Ore:	Portland	3	3	24	23	0	0	111	98
Pa:	Philadelphia	3	3	17	17	0	0	74	68
	Pittsburgh	3	4	25	24	2	2	101	94
P. R:	San Juan	3	3	11	11	1	1	56	53
R. I:	Providence	3	3	20	19	0	0	108	102
S. C:	Charleston	3	3	29	29	1	1	98	95
S. Dak:	Rapid City	5	7	33	30	3	4	119	110
Tenn:	Chattanooga	4	4	33	32	1	1	71	67
	Memphis	3	4	26	25	2	2	46	44
Tex:	Austin	3	3	8	8	1	2	29	29
	Dallas	3	3	18	17	1	1	42	40
Utah:	Salt Lake City	3	3	23	22	2	2	126	117
Vt:	Burlington	3	3	22	21	2	2	118	111
Va:	Norfolk	4	4	16	16	2	3	64	60
Wash:	Seattle	3	3	24	23	0	0	115	104
	Spokane	3	3	25	25	0	0	104	97
W. Va:	Charleston	4	4	19	19	2	2	45	43
Wis:	Milwaukee	3	3	15	14	3	3	90	85
Wyo:	Laramie	3	3	16	15	2	2	77	71
Network average		3	3	20.3	19.6	2	2	80	75

[a] Annual averages for barium-140 at each station were <2 pCi/liter.
[b] Annual averages were computed on basis of 48 weekly averages.

(2) NATIONAL COMMITTEE ON RADIATION PROTECTION. Maximum permissible body burdens and maximum permissible concentrations of radionuclides in air and in water for occupational exposure. National Bureau of Standards Handbook 69. Superintendent of Documents, U.S. Government Printing Office, Washington, D. C. 20402 (June 5, 1959).

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CESIUM–137 IN FRESHWATER FISH: HUMAN CONSUMPTION AND BODY BURDEN CONSIDERATIONS[1]

P. F. Gustafson[2]

Measurements of cesium–137 in freshwater fish made at Argonne National Laboratory during the first half of 1965 indicated relatively high concentrations (*1*). The fish examined were purchased in food markets in the Chicago area and, for the most part, were fresh or frozen dressed ready for cooking. Frozen fish may be kept for a considerable time without spoilage; hence, those samples investigated may have included fish actually caught during the 1964 season, or perhaps even earlier. The work of Häsänen and Miettinen in Finland showed comparable or slightly lower levels in freshwater fish caught during 1961 and 1962 (*2*). More recently Kolehmainen *et al.* reported increased concentrations in fish from the same lakes during 1963 and early 1964 (*3*). Analyses of the Chicago portion of the foods collected as part of the Tri-City Diet Sampling Program showed exceptionally high cesium–137 concentrations in fish on several occasions prior to 1965 (*4*). The fish purchased in the Tri-City study were usually ocean varieties, with ocean perch, cod, and halibut predominating. When freshwater fish were included, they were mixed with the other varieties and the homogenized batch of fish was measured for cesium–137. Thus the present observation of high cesium–137 levels in freshwater fish need not be considered a sudden or recent occurrence; nor is there any reason to presume it to be a local phenomenon. Due to the efficient processing and distribution of fish and fish products, freshwater fish containing similar concentrations of cesium–137 should be readily available on the commercial market throughout most of the United States and Canada.

Cesium–137 concentrations found in several of the more important freshwater species ranged from 180 pCi/kg (wet weight) in whitefish, to more than 4,000 pCi/kg in some of the larger predators such as lake trout and pike, as shown in table 1. For comparison, the levels seen in a number of marine species, ranging from 10 to approximately 100 pCi/kg, are also listed in table 1. Although not all commercially important varieties of freshwater fish have as yet been examined, table 1 clearly indicates that in every case thus far, the concentration of cesium–137 was substantially greater in freshwater than in marine species.

Table 1. Range of cesium–137 and potassium concentrations and $^{137}Cs/K$ ratio observed in freshwater and ocean fish purchased during January to August 1965

Species[1]	^{137}Cs (pCi/kg wet weight)	Potassium (g/kg wet weight)	$^{137}Cs/K$ ratio (pCi/g)
Freshwater fish			
Lake trout	2,210–4,230	2.49–3.50	890–1,230
Northern pike	1,170–4,100	2.47–3.58	440–1,210
Whitefish	180–2,800	2.68–3.77	55–800
Lake perch	330–1,105	2.96–3.40	110–390
Chubs[2]	680–1,400	3.10–3.50	220–430
Smelts	325–430	2.60–3.30	125–160
Ocean fish			
Cod	29–43	3.90–4.17	7–10
Haddock	53–66	3.70–4.02	14–16
Halibut	62–69	5.26–5.35	12–13
Flounder	24–39	2.88–3.10	8–12
Ocean perch	32–56	3.08–3.11	10–18
Ocean catfish	43–59	4.04–4.15	11–14
Red snapper	12–21	1.76–1.95	7–11
Salmon[3]	71–93	3.51–3.66	20–25

[1] The common or commercial names of fish are used here instead of the scientific (taxonomic) names because of the difficulty in determining the latter with any precision in the case of frozen, dressed fish.
[2] Smoked chubs were analyzed.
[3] Salmon also contained concentrations of manganese-54 comparable to that of cesium-137.

The capacity of various members of a freshwater aquatic system to concentrate cesium–137 to a high degree was illustrated in the work of Pendleton and Hanson in 1958, when they investigated a number of species of aquatic animals living in a freshwater pond contaminated with cesium–137 (*5*). The most important factor governing the uptake of cesium–137 in going from one trophic level to another appears to be the $^{137}Cs/K$ ratio itself. Because of the higher concentration of potassium in sea water (approximately 380 parts per million) relative to that in fresh water (a few parts per million), equal concentrations of cesium–137 in the two media will result in a higher $^{137}Cs/K$ ratio in fresh water. Pendleton *et al.* have ob-

[1] Work supported by the U.S. Atomic Energy Commission.
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served on the order of a threefold increase in the $^{137}Cs/K$ ratio in proceeding from one trophic level to the next higher level in both freshwater and terrestrial environments (*6*). These authors attribute the increase in ratio to the longer biological half-life of cesium relative to potassium. Thus, the cesium–137 concentration in a specific fish should be a function of the $^{137}Cs/K$ ratio in the water and the position of the fish on the trophic scale.

Relative consumption of fish

The immediate interest in the cesium–137 levels in freshwater fish relates to the concentration of cesium–137 in the human body which may result from the eating of such fish. In general, the U.S. population is not heavily dependent upon aquatic food (*i.e.*, fish) as a source of protein. This is illustrated by statistics for 1964 which showed that the average annual per capita consumption of fish in the United States was 4.8 kg, placing it fourth (in terms of mass consumed) in dietary importance as a protein source after milk (187 kg), meat (77 kg), and poultry (including turkey, 17 kg) (*7*). The bulk of fish eaten in this country come from the ocean; of the nearly one billion kg of fish actually consumed by man in the United States in 1964, only 4 to 5 percent (making some allowance for fish caught by sport fishermen) were freshwater fish (*8*). It should be emphasized, however, that the total gross weight of fish caught by U.S. fishermen, and imported, amounted to 5.7 billion kg in 1964 (*8*). The 4.6 billion kg of nonedible fish and fish products stayed nonetheless within the broader confines of the food chain, since fish meal is used as an animal and poultry feed, and fish fertilizer is used in agriculture.

Cesium–137 intake from fish

The cesium–137 level in the composite Chicago diet has been determined quarterly since April 1961 as part of the Argonne contribution to the Tri-City program. In April 1965, the adult intake of cesium–137 according to this diet was 140 pCi/day contained in 1.85 kg of food. In this particular sampling, the fish portion of the diet consisted of halibut, cod, haddock, sole, ocean perch, pike, trout, and whitefish, the latter three species being from fresh water. The arithmetic average concentration of cesium–137 in all fish contributes a daily intake of 8 pCi/day, assuming an annual consumption of 8 kg of fish. The excess over the national average, 8 kg versus 4.8 kg, is presumably due to the proximity of New York City, Chicago, and San Francisco to an abundant supply of fish. By using the April 1965 concentrations of cesium–137 instead of taking the arithmetic average, assuming that 5 percent of the daily intake came from freshwater fish (average concentration of 930 pCi/kg), and the other 95 percent came from ocean fish (average concentration of 27 pCi/kg), then the daily intake due to fish would be less than 2 pCi/day.

The important fact to bear in mind, however, is that the above are average figures, and some population groups may eat substantially larger amounts of fish. There are also individual persons who eat fish in preference to meat or poultry. In addition to such dietary preferences, one must consider religious, cultural, and health factors which may also cause an increase above the national average. Proximity to an abundant supply of fish has already been cited as a reason for increased intake of cesium. Further influence in this direction can be visualized in remote areas where fishing is the principal means of livelihood. Of particular interest in regard to cesium–137 would be fishing camps and processing plants located along freshwater lakes and streams in remote localities.

Changes in the diet can be made which will produce an increase in freshwater fish consumption, and in turn may cause an increase in the cesium–137 whole-body burden. The relationship between the level of cesium–137 in the diet and that in man, under near equilibrium conditions, has been investigated using the Tri-City diet results and whole-body counter measurements made at Argonne. The average cesium–137 intake for Chicago adults during the first 9 months of 1964 was computed to be 201.5 pCi/day, and the corresponding average potassium intake was 4.5 g/day, resulting in an average ratio in the entire diet of 44.8 pCi/g K. The average daily potassium intake observed during 1962 through 1964 was 4.5 ± 0.39, and a value of 4.5 g/day will be used in all subsequent calculations of $^{137}Cs/K$ values. In September 1964 whole-body measurements by Miller showed an average $^{137}Cs/K$ ratio of 139.8 pCi/g K in 8 Argonne employees (*9*).

The increase ratio shown in equation 1,

$$\frac{(^{137}\text{Cs/K})\ \text{man}}{(^{137}\text{Cs/K})\ \text{diet}} = \frac{139.8}{44.8} = 3.12, \qquad \text{Eq. 1}$$

was 3.12, in accord with the findings of Pendleton *et al.* (*6*), referred to previously. This would seem to indicate that the factor of 3 increase in ^{137}Cs/K ratio applies also in the case of man relative to his diet, and will be used in subsequent calculations.

As mentioned previously, in April 1965 the cesium–137 content of the composite adult diet in the Chicago area was 140 pCi/day; this, coupled with a potassium intake of 4.5 g, yields a ^{137}Cs/K ratio of 31.1 pCi ^{137}Cs/g K. The cesium–137 and potassium intakes from various categories of foodstuffs, as well as the ^{137}Cs/K ratio for each component, are shown in table 2. Milk, meat, and grain products account for 80 percent of the daily intake of cesium–137 and nearly 50 percent of the daily intake of potassium. Even in the case of mixed fish, the ^{137}Cs/K ratio in fish is higher than any of the other diet components, being followed by grain products, meat, and milk, in that order.

Table 2. Cesium-137, potassium, and ^{137}Cs/K ratio in the adult Chicago diet, April 1965

Diet component	Cesium-137 (pCi/day)	Potassium (g/day)	^{137}Cs/K, (pCi/g K)
Milk	47.2	1.09	43.3
Meat	36.2	0.63	57.5
Grain products	31.7	0.43	73.7
Fruit	11.1	0.66	16.8
Fish (mixed)	8.0	0.09	88.9
Vegetables	3.6	1.39	2.6
Poultry	1.5	0.12	12.5
Eggs	0.8	0.07	11.4
Total	140.1	4.48	*31.2

* Weighted total on the basis of the amount of each component consumed per day.

The ^{137}Cs/K ratio in man at or near equilibrium with a diet containing a ratio of 31.1 pCi/g K would be approximately 100 pCi/g K. Assuming a standard 70 kg man to contain 140 g of potassium, a level of 100 pCi/g K corresponds to a total cesium–137 body burden of 14,000 pCi or 14 nCi (nanocuries).[3] This cesium–137 body burden is in accord with whole-body measurements made slightly prior to this time (*10*).

[3] Editor's note: For reference, NBS Handbook 69 (*13*) lists the following Maximum Permissible Body Burdens of ^{137}Cs:
For radiation workers (occupational), 30 μCi.
For individuals in the general population, 3 μCi.
For the average person in the general population, 1 μCi.

Consider an individual who divides the protein intake (81 kg/year) assumed to come from fish (8 kg/year) and meat (73 kg/year) equally between fish and meat, other diet components remaining the same. If all the fish were from fresh water, the cesium–137 intake would become 218 pCi/day, and the ^{137}Cs/K ratio would be 48.8 pCi/g. Limiting the entire protein intake of 81 kg/year to freshwater fish would raise the cesium–137 intake to 302 pCi/day, with a ^{137}Cs/K ratio of 67.5 pCi/g, which would lead to a body burden of 28 nCi at equilibrium, or double the observed level in the Chicago population in April 1965.

An annual consumption of 81 kilograms of meat and fish amounts to slightly under a half-pound of protein per day. People on a less varied diet, surrounded by an ample supply of fish and little else, might be expected to eat more than a half-pound of fish per day. Miettinen *et al.*, in studying the Lapland diet, found that the male Lapp eats on the average more than 500 grams of meat and fish per day (*11*). It should also be borne in mind that the concentration of 930 pCi/kg of wet weight observed in freshwater fish in April 1965 is surpassed in a number of instances, as shown in table 1. The mean value of several dozen northern pike from Red Lake, Minnesota, measured at Argonne was approximately 3,000 pCi/kg (approximately 1,000 pCi/g K). Fish at this cesium–137 level eaten in the amount of *1 pound per day* would lead to an equilibrium body burden in the standard man of 420 nCi. Cesium–137 concentrations in freshwater fish, higher by factors of 3 to 6, have been observed by Canadian and Finnish investigators during 1964 and 1965 (*3, 12*).

Evidence of the importance of freshwater fish as a dietary source of cesium–137 has come from the studies of Lapps and Eskimos. These people obtain much of their cesium–137 from eating caribou and/or reindeer. However, fish are also plentiful and, more importantly, available on a year-round basis. Miettinen found in 1962 that 10–15 percent of the cesium–137 intake in Lapps came from fish (no differentiation between freshwater and ocean fish was made) at a time when the whole-body burdens were 200–400 nCi, thus implying that 20 to 60 nCi came from fish (*11*). In 1964 Bird *et al.* noted two segments of the Canadian Eskimo population whose body burdens were several hundred nCi but who ate little or no caribou or

reindeer (*14*). It would thus appear that even in the case of persons eating caribou and reindeer, freshwater fish are making a substantial contribution to the total body cesium–137 burden, even though the levels of cesium–137 in fish observed thus far are lower than those found in caribou and reindeer.

There is thus strong suggestive evidence that body burdens of the order of 0.1 μCi (100 nCi) could be attained by people not dependent upon caribou or reindeer for food, but subsisting to a large extent upon fish and other natural forms such as deer or other mammals which might also be expected to have substantial concentrations of cesium–137. Interest in these fish-eating populations should approach that being given to the Lapps and Eskimos, particularly since their number may well exceed that in the caribou-reindeer eating population. The fish-eaters thus comprise a potentially important subgroup of the population and more information about them should be obtained.

Having established the existence of relatively high cesium–137 levels in freshwater fish, a further question is, how do these levels change with time, and in particular, what future levels may we expect? The biologic half-time of cesium in fish is short (5–10 days) (*15*); thus, changes in the cesium–137 content in the diet of the fish will cause rapid changes in the body burden of fish. There are seasonal changes in the eating habits of fish, and there are also changes in the deposition of cesium–137 throughout the year. Analyses of fish specimens collected over past years—particularly the crucial years 1957–1964—would be invaluable in this regard. Such data used in conjunction with cesium–137 deposition and accumulation data could establish future trends on the basis of past behavior.

The presence of readily measurable quantities of cesium–137 in freshwater fish makes it possible to conduct a variety of investigations of environmental contamination, not all of which relate solely to radioactive contamination. The most obvious opportunity is probably that of studying the freshwater ecosystem itself. Some groups in the fish-eating population offer perhaps an unmatched opportunity to study a remote, nonmobile population largely unaffected by the averaging and dilution processes inherent in modern food technology, yet very strongly affected by the surrounding environmental factors. A detailed and imaginative investigation could provide the basis for the sound evaluation of the mechanisms and effects of localized (in the broad sense) contamination—a matter of current and lasting concern to radiation surveillance and protection groups. Furthermore, such an investigation, involving a variety of disciplines, could yield much basic information on the movement, uptake, retention, and elimination of contaminants in a real situation which could not be duplicated in the laboratory.

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Section III—Water

GROSS RADIOACTIVITY IN SURFACE WATERS OF THE UNITED STATES, MAY 1965

Division of Water Supply and Pollution Control Public Health Service

Levels of radioactivity in surface waters of the United States have been monitored by the Public Health Service Water Pollution Surveillance System since its initiation in 1957. Beginning with the establishment of 50 sampling points, this system has been expanded to include 131 stations. These stations are operated jointly with other Federal, State, and local agencies, and with private industries. Samples are taken from surface waters of all major U.S. river basins for physical, chemical, biological, and radiological analyses. The system provides background information necessary for recognizing water quality trends and for determining current and general levels of surface water contamination and early detection of specific situations which may warrant more detailed evaluation. Data assembled through the system and exact locations of sampling points are published in annual compilations (*1–8*).

Sampling procedures

The participating agencies collect 1-liter "grab" samples each week and ship them "as is" to the Surveillance System Laboratory in Cincinnati for analysis. Gross alpha and gross beta radioactivity determinations on the suspended and dissolved solids are performed as frequently as deemed necessary. Presently, gross alpha and beta determinations are made either on monthly composites of the weekly samples or on each weekly sample. Weekly alpha and beta determinations are scheduled for stations located downstream from known potential sources of radioactive waste. Weekly analyses are conducted at all newly established stations for the first year of operation. Weekly analyses are also scheduled for selected stations in an effort to detect short term increases in radioactivity from current or recent nuclear tests or events.

Analytical methods

The analytical method for determining gross alpha and beta radioactivity is described in the eleventh edition of "Standard Methods for the Examination of Water and Wastewater" (*9*). Suspended and dissolved solids are separated by passing the sample through a membrane filter (type HA) with a pore size of 0.45 micron. Planchets are then prepared for counting the dissolved solids (in the filtrate) and the suspended solids (on the charred membrane filter) in an internal proportional counter. Reference sources of U_3O_8, which give a known count rate if the instrument is performing properly, are used for daily checking of the counter.

Normally, samples are counted within 2 weeks following collection or within 1 week after compositing. The decay of activity is followed for each sample for which the first analysis shows unusually high activity. Also, if a recount indicates that the original analysis was questionable, values based on the recount are recorded. All results are reported for the time of counting and are not extrapolated to the date of collection.

Results

Table 1 presents the most recent results of alpha and beta analysis of U.S. surface waters. The stations on a river are arranged in the table according to their relative locations, the first stations listed being closest to the head-

Table 1. Radioactivity in raw surface waters, May 1965[a]

(Average concentrations in pCi/liter)

Station	Beta activity			Alpha activity		
	Suspended	Dissolved	Total	Suspended	Dissolved	Total
Allegheny River:						
Pittsburgh, Pa	1	5	6	0	0	0
Animas River:						
Cedar Hill, N. Mex	7	7	14	1	1	2
Apalachicola River:						
Chattahoochee, Fla	—	—	—	—	—	—
Arkansas River:						
Coolidge, Kans	273	37	310	96	23	119
Ponca City, Okla	37	16	53	13	3	16
Fort Smith, Ark	15	18	33	5	2	7
Little Rock, Ark	219	15	234	30	1	31
Pendleton Ferry, Ark	8	15	23	2	2	4
Atchafalaya River:						
Morgan City, La	48	11	59	20	1	21
Bear River:						
Preston, Idaho	2	12	14	1	2	3
Big Horn River:						
Hardin, Mont	69	20	89	13	7	20
Big Sioux River:						
Sioux Falls, S. Dak	7	33	40	1	5	6
Chattahoochee River:						
Atlanta, Ga	—	—	—	—	—	—
Lanett, Ala	2	4	6	0	0	0
Columbus, Ga	0	4	4	0	0	0
Chena River:						
Fairbanks, Alaska	5	9	14	1	1	2
Clearwater River:						
Lewiston, Idaho	3	3	6	<1	0	<1
Clinch River:						
Clinton, Tenn	1	3	4	0	0	0
Kingston, Tenn	—	—	—	—	—	—
Colorado River:						
Loma, Colorado	114	15	129	123	3	126
Page, Ariz	2	25	27	0	9	9
Boulder City, Nev	—	—	—	—	—	—
Parker Dam, Calif-Ariz	2	22	24	0	9	9
Yuma, Ariz	1	31	32	0	0	0
Columbia River:						
Northport, Wash	1	4	5	0	1	1
Wenatchee, Wash	1	5	6	0	1	1
Pasco, Wash	21	105	126	0	1	1
McNary Dam, Ore	37	96	133	1	0	1
Bonneville, Ore	—	—	—	—	—	—
Clatskanie, Ore	90	48	138	1	0	1
Connecticut River:						
Wilder, Vt	1	5	6	0	0	0
Northfield, Mass	24	4	28	4	0	4
Enfield Dam, Conn	2	5	7	1	0	1
Coosa River:						
Rome, Ga	2	6	8	<1	0	<1
Cumberland River:						
Cheatham Lock, Tenn	0	5	5	1	0	1
Cuyahoga River:						
Cleveland, Ohio	3	23	26	1	1	2
Delaware River:						
Martins Creek, Pa	1	12	13	0	0	0
Trenton, N.J	1	5	6	0	0	0
Philadelphia, Pa	1	4	5	0	0	0
Escambia River:						
Century, Fla	1	4	5	0	0	0
Great Lakes:						
Duluth, Minn	1	3	4	0	0	0
Sault Ste. Marie, Mich	4	5	9	0	0	0
Milwaukee, Wis	0	6	6	0	0	0
Gary, Ind	1	6	7	0	0	0
Port Huron, Mich	0	8	8	0	0	0
Detroit, Mich	0	6	6	0	0	0
Buffalo, N.Y	—	—	—	—	—	—
Green River:						
Dutch John, Utah	1	22	23	0	4	4
Hudson River:						
Poughkeepsie, N.Y	2	6	8	0	0	0
Illinois River:						
Peoria, Ill	12	15	27	3	2	5
Grafton, Ill	—	—	—	—	—	—
Kanawha River:						
Winfield Dam, W. Va	1	5	6	0	0	0
Kansas River:						
De Soto, Kans	7	15	22	2	3	5
Klamath River:						
Keno, Ore	1	17	18	0	1	1
Little Miami River:						
Cincinnati, Ohio	2	7	9	0	0	0
Maumee River:						
Toledo, Ohio	5	16	21	1	2	3
Merrimack River:						
Lowell, Mass	—	—	—	—	—	—
Mississippi River:						
St. Paul, Minn	5	29	34	1	4	5
Dubuque, Iowa	5	15	20	1	1	2
Burlington, Iowa	4	16	20	1	1	2
E. St. Louis, Ill	11	16	27	2	3	5
Cape Girardeau, Mo	—	—	—	—	—	—
W. Memphis, Ark	5	16	21	1	1	2
Vicksburg, Miss	13	12	25	3	0	3
Delta, La	16	13	29	5	1	6
New Roads, La	10	11	21	3	0	3
New Orleans, La	12	12	24	5	1	6
Missouri River:						
Williston, N. Dak	7	18	25	2	4	6
Bismarck, N. Dak	16	20	36	6	4	10
Yankton, S. Dak	2	27	29	0	6	6
Omaha, Nebr	26	19	45	7	4	11
St. Joseph, Mo	71	20	91	18	3	21
Kansas City, Kans	14	24	38	5	5	10
Missouri City, Mo	—	—	—	—	—	—
St. Louis, Mo	30	23	53	7	2	9
Monongahela River:						
Pittsburgh, Pa	1	5	6	0	1	1
North Platte River:						
Henry, Nebr	11	32	43	1	22	23
Ohio River:						
Toronto, Ohio	1	8	9	0	0	0
Addison, Ohio	—	—	—	—	—	—
Huntington, W. Va	—	—	—	—	—	—
Cincinnati, Ohio	0	5	5	0	0	0
Louisville, Ky	3	6	9	0	1	1
Evansville, Ind	9	7	16	2	0	2
Cairo, Ill	7	8	15	1	1	2
Ouachita River:						
Bastrop, La	—	—	—	—	—	—
Pend Oreille River:						
Albeni Falls Dam, Idaho	3	4	7	0	<1	<1
Platte River:						
Plattsmouth, Nebr	307	22	329	79	3	82
Potomac River:						
Williamsport, Md	1	3	4	0	0	0
Great Falls, Md	1	5	6	0	0	0
Washington, D. C	1	4	5	0	0	0
Rainy River:						
Baudette, Minn	3	20	23	0	<1	<1
International Falls, Minn	2	18	20	0	0	0
Raritan River:						
Perth Amboy, N. J	—	—	—	—	—	—
Red River, North:						
Grand Forks, N. Dak	12	35	47	3	3	6
Red River, South:						
Denison, Tex	1	24	25	0	2	2
Index, Ark	—	—	—	—	—	—
Bossier City, La	2	17	19	0	3	3
Alexandria, La	16	15	31	7	1	8
Rio Grande:						
Alamosa, Colo	—	—	—	—	—	—
El Paso, Tex	6	18	24	1	5	6
Laredo, Tex	46	15	61	10	2	12
Brownsville, Tex	—	—	—	—	—	—
Roanoke River:						
John H. Kerr Resr. & Dam, Va	1	5	6	0	0	0
Sabine River:						
Ruliff, Texas	6	12	18	1	0	1
Sacramento River:						
Greens Landing, Calif	—	—	—	—	—	—
St. Lawrence River:						
Massena, N. Y	3	8	11	0	0	0
San Joaquin River:						
Vernalis, Calif	4	6	10	1	0	1
San Juan River:						
Shiprock, N. Mex	25	11	36	8	2	10
Savannah River:						
North Augusta, S. C	2	6	8	0	0	0
Port Wentworth, Ga	3	8	11	<1	0	<1
Schuylkill River:						
Philadelphia, Pa	1	9	10	0	0	0
Shenandoah River:						
Berryville, Va	—	—	—	—	—	—
Ship Creek:						
Anchorage, Alaska	—	—	—	—	—	—
Snake River:						
Payette, Idaho	3	7	10	1	1	2
Wawawai, Wash	3	4	7	<1	0	<1
Ice Harbor Dam, Wash	1	4	5	0	0	0
South Platte River:						
Julesburg, Colo	88	58	146	18	36	54

See footnote at end of table.

Table 1. Radioactivity in raw surface waters, May 1965[a]—Continued

Average concentrations in pCi/liter

Station	Beta activity			Alpha activity		
	Suspended	Dissolved	Total	Suspended	Dissolved	Total
Spokane River:						
Post Falls Dam, Idaho	1	2	3	0	0	0
Susquehanna River:						
Sayre, Pa	1	10	11	0	0	0
Conowingo, Md	1	3	4	0	0	0
Tennessee River:						
Lenoir City, Tenn	1	4	5	0	0	0
Chattanooga, Tenn	1	10	11	0	0	0
Bridgeport, Ala	—	—	—	—	—	—
Pickwick Landing, Tenn	1	7	8	0	0	0
Tombigbee River:						
Columbus, Miss	2	5	7	1	0	1
Truckee River:						
Farad, Calif	0	4	4	0	0	0
Verdigris River:						
Nowata, Okla	173	16	189	54	0	54
Wabash River:						
New Harmony, Ind	16	10	26	3	1	4
Willamette River:						
Portland, Ore	1	2	3	0	0	0
Yakima River:						
Richland, Wash	1	4	5	0	0	0
Yellowstone River:						
Sidney, Mont	52	15	67	16	3	19
Maximum	307	105	329	123	36	126
Minimum	0	2	3	0	0	0

[a] These data are preliminary: reanalysis of some samples may be made and additional analyses not completed at the time of this report may become available. For final data, one should consult the system's annual report.

Dashes indicate no sample.

waters. These data are preliminary. The figures for gross alpha and gross beta radioactivity represent either determinations on composite samples or means of weekly determinations where composites were not made. The monthly means are reported to the nearest pCi/liter. When all samples have zero pCi/liter, the mean is reported as zero; when the calculated mean is between zero and 0.5, the mean is reported as <1 pCi/liter.

Figure 1. Sampling locations and associated total beta activity (pCi/liter) in surface waters, May 1965

It has been observed that in water the natural environmental beta activity is usually several times that of the natural environmental alpha activity. Nuclear installations may contribute additional alpha and/or beta activity whereas fallout primarily contributes additional beta activity.

The radioactivity associated with dissolved solids provides a rough indication of the levels which would occur in treated water, since nearly all suspended matter is removed by treatment processes (*10*). The Public Health Service Drinking Water Standards state that in the absence of alpha emitters and strontium–90, a water supply is acceptable when the gross beta concentration does not exceed 1,000 pCi/liter (*11*).[1]

A geographical perspective of the radioactivity in surface water is obtained from the numbers printed near the stations as shown in figure 1, which gives the average total beta activity in suspended-plus-dissolved solids in raw water collected at each station.

Table 2. Summary

Station	Sample activity	Remarks
Arkansas River Coolidge, Kans.	α, Susp. 384 pCi/liter β, Susp. 1086 pCi/liter on sample collected 5/24/65, analyzed in triplicate; median reported.	Due to flood condition, sample contained 14,930 mg/l suspended solids.
Little Rock, Ark.	α, Susp. 30 pCi/liter β, Susp. 219 pCi/liter	Sample contained 4,288 mg/l suspended solids.
Colorado River Loma, Colo.	α, Susp. 123 pCi/liter β, Susp. 114 pCi/liter	Sample contained only 720 mg/l suspended solids. Level returned to normal <10 pCi/l on next sample.
Missouri River St. Joseph, Mo.	α, Susp. 18 pCi/liter β, Susp. 71 pCi/liter	Sample contained 1,002 mg/l suspended solids.
Platte River Plattsmouth, Neb.	Average for month α, Susp. 79 pCi/liter β, Susp. 307 pCi/liter	Average for month contained 5,128 mg/l suspended solids.
Verdigris River Nowata, Okla.	α, Susp. 54 pCi/liter β, Susp. 173 pCi/liter Analyzed in triplicate; median reported.	Sample contained 2,304 mg/l suspended solids.

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(*11*) PUBLIC HEALTH SERVICE. Drinking water standards, revised 1962. Public Health Service Publication No. 956. Superintendent of Documents, U.S. Government Printing Office, Washington, D.C. 20402 (March 1963). Price 30 cents.

[1] Absence is taken here to mean a negligibly small fraction of the specific limits of 3 pCi/liter and 10 pCi/liter for alpha emitters and strontium–90, respectively.

[2] Single free copies of this publication may be obtained from Public Inquiries Branch, Public Health Service, U.S. Department of Health, Education, and Welfare, Washington, D.C. 20201.

RADIOACTIVITY IN SURFACE WATERS OF THE COLORADO RIVER BASIN, 1962–1964

Colorado River Basin Water Quality Control Project
Division of Water Supply & Pollution Control, PHS

In view of accumulated data on radioactive contamination of Colorado River Basin surface waters and river sediments by uranium milling waste discharges to the stream environment, a conference on uranium milling wastes was held in October 1957 at Cortez, Colorado. The conference was attended by representatives of the States of Arizona, New Mexico, Colorado, Utah, and the U.S. Public Health Service. At this conference, a Radium Monitoring Network was suggested to provide needed continuous data on radioactivity contributed by uranium mining and milling operations to the Colorado River Basin surface waters. The envisioned purpose of this network was to define both natural and industrial sources of radioactive pollution, and to measure either improvement or degradation of surface waters due to the continuance or abatement of these pollution sources. Accordingly, a Radium Monitoring Network was established when the U.S. Public Health Service Colorado River Basin Water Quality Control Project was organized in 1960 under provisions of the Federal Water Pollution Control Act. The original network of 23 stations became operational at the end of 1961, was expanded to 27 stations (figure 1), and has recently been reduced to 21 active stations. Data collected through this system are reported seminannually to the Project conferees and other interested parties and agencies, and a summary of data collected through 1963 was published in November 1964 (*1*).

Sampling procedures

Seven of the original network stations were automatic collection stations and the remainder were grab sampling stations. Depending upon the type of data required and physical location limitations encountered at each station, grab samples are collected 1 to 6 times weekly and automatic samplers collect a 21-ml sample every hour. The samples are sent "as is" to the Colorado River Basin Project Laboratory where they are composited into weekly or monthly composite samples, filtered, and the filtrate analyzed for the desired radionuclides.

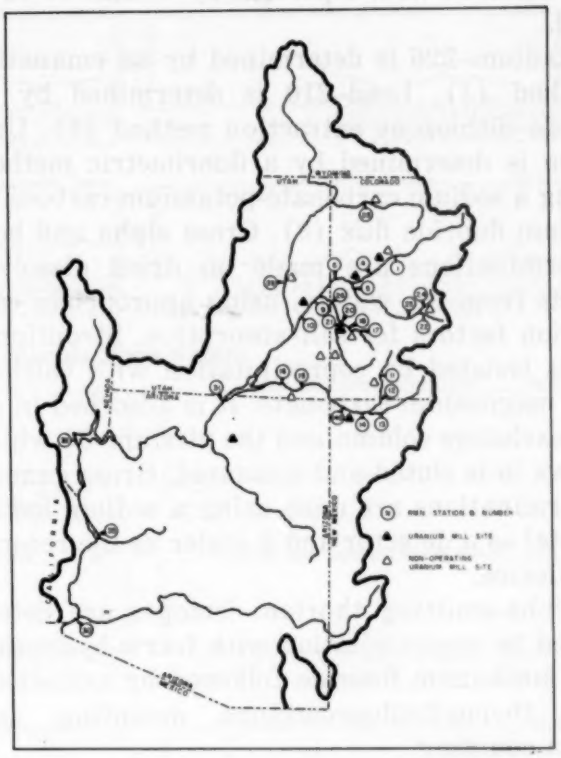

Figure 1. Colorado River Basin radium monitoring network

Radium–226, with the lowest recommended Maximum Permissible Concentration in water of all radionuclides (*2*), has been the radioactive contaminant of greatest concern in uranium mill discharges (*3*). Consequently, initial and primary emphasis was placed on the analysis for radium–226 only. In October 1963, uranium determinations were added, and beginning in July 1963, samples from selected stations were combined into quarterly composites to be gamma-scanned and analyzed for gross alpha, gross beta, lead–210, thorium alpha, and strontium–90 activities. Some "spot check" determinations for polonium–210 have also been performed. Additional analysis for this radionuclide is required before any data can be reported.

These determinations have been added to provide a more definitive picture of the total radioactivity in Basin surface waters resulting from uranium industry operations and nuclear weapons–testing programs.

Analytical methods

Water samples are composited and filtered through 0.45–micron membrane filters as soon as possible after the samples for the compositing period are received. After filtration they are acidified with 2 percent by volume of 12 *N* HCl.

Radium–226 is determined by an emanation method (*4*). Lead–210 is determined by an iodide–dithiozone extraction method (*5*). Uranium is determined by a fluorimetric method, using a sodium carbonate-potassium carbonate-sodium fluoride flux (*6*). Gross alpha and beta determinations are made on dried dissolved solids from the waters, using appropriate correction factors for self-absorption. Strontium–90 is isolated by coprecipitation with calcium and magnesium carbonate. It is adsorbed in an ion exchange column and the yttrium–90 which grows in is eluted and measured. Gross gamma determinations are done using a sodium iodide crystal as a detector and a scaler as the recording device.

Alpha-emitting thorium isotopes are determined by coprecipitation with ferric hydroxide and lanthanum fluoride followed by extraction with thenoyltrifluoroacetone, mounting, and alpha counting.

Results

A summary of all radium–226 and uranium determinations on Radium Monitoring Network samples is given in table 1. The figures given in the table represent all sampling results obtained since the inception of the first network stations in October and November 1961. Figure 2 presents a graphical summary of radium–226 results at each individual station and table 2 gives the uranium data. Table 3 presents the quarterly composite results for the gross alpha, gross beta, lead–210, thorium alpha, and strontium–90. These latter results cover the period July 1963 through June 1964. Results on all reported parameters are for the dissolved solids portion of the sample passing the 0.45 micron filter.

In keeping with the philosophy of minimum exposure (*2*), the majority of the uranium processing mills in the Colorado River Basin have successfully maintained negligible radioactivity contributions to Colorado River Basin waters through their efforts to provide proper treatment and containment of milling waste products. Even at those mills where significant contributions have occurred since 1960, only eight of the 1,410 composite samples analyzed for radium–226 have indicated concentrations

Table 1. Radium–226 and uranium in surface waters of the Colorado River Basin

Sampling station number and location	Radium-226			Uranium		
	Average (pg/liter)	Range (pg/liter)	No. of analyses	Average (ug/liter)	Range (ug/liter)	No. of analyses
Animas River:						
#11 above Durango, Colo	0.05	0.03–0.11	38	1.1	0.1–2.5	32
#12 Colorado-New Mexico State line	0.30	0.09–1.5	89	4.0	0.4–18	47
Colorado River:						
# 1 at Silt, Colo	0.21	0.04–0.39	43	3.0	0.1–4.4	29
# 4 at DeBeque, Colo	0.20	0.05–0.44	44	3.8	0.4–7.2	29
# 6 at Fruita, Colo	0.19	0.09–0.26	39	9.7	3.2–14	30
# 9 above Moab, Utah	0.27	0.10–0.50	72	10	3.6–17	59
#10 below Moab, Utah	0.43	0.18–7.2	68	12	3.7–22	55
#31 at Page, Ariz	0.24	0.16–0.31	22	8.8	6.5–13	22
#32 Lake Mead, Nev	0.32	0.21–0.40	24	8.5	5.6–11	24
#33 Lake Havasu, Calif	0.35	0.06–0.41	26	8.4	5.9–11	26
#30 at Yuma, Ariz	0.17	0.06–0.33	34	8.3	5.9–11	29
Dolores River:						
#21 at Bedrock, Colo	0.51	0.03–1.4	42	14	0.9–35	29
#26 at Gateway, Colo	1.2	0.04–3.2	81	13	0.1–51	58
Green River:						
#28 below Maybell, Colo	0.09	0.03–0.15	33	2.7	0.6–8.0	29
#29 below Green River, Utah	0.09	0.05–0.15	28	5.0	1.6–7.6	25
Tomichi Creek:						
#22 above Gunnison, Colo	0.02	0.01–0.04	7	NA	NA	NA
Gunnison River:						
#24 above Gunnison, Colo	0.05	0.01–0.08	39	1.2	0.0–2.6	29
#25 below Gunnison, Colo	0.04	0.01–0.09	42	2.2	0.6–4.3	29
# 5 at Grand Junction, Colo	0.10	0.01–0.30	38	11	0.7–19	29
San Juan River:						
#13 above Farmington, N. Mex	0.07	0.03–0.16	20	2.8	0.0–8.3	19
#14 below Farmington, N. Mex	0.10	0.04–0.21	30	3.6	0.9–9.0	29
#14S below Shiprock, N. Mex	0.10	0.04–0.26	95	14	2.0–93	91
#15 above Mexican Hat, Utah	0.14	0.04–0.66	42	7.8	2.6–23	29
#16 below Mexican Hat, Utah	0.14	0.01–0.84	113	9.8	0.8–35	95
San Miguel River:						
#17 above Naturita, Colo	0.04	0.00–0.19	57	1.2	0.0–3.9	29
#18 above Uravan, Colo	0.23	0.05–1.24	90	6.8	0.9–63	59
#20 below Uravan, Colo	1.0	0.06–5.6	154	31	0.0–150	126

NA indicates no analysis performed.

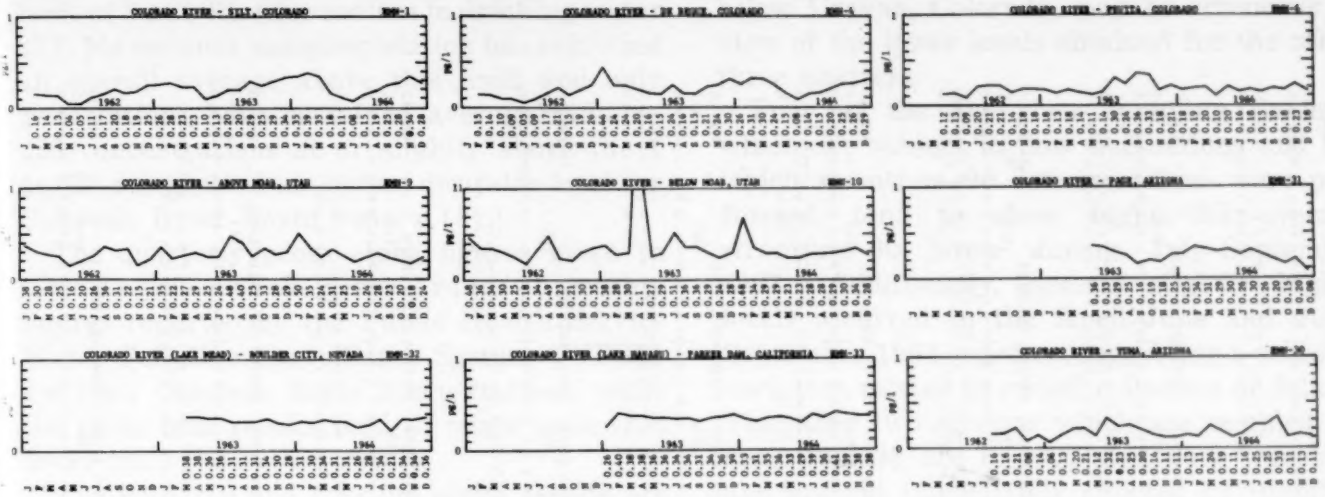
COLORADO RIVER - SILT, COLORADO
COLORADO RIVER - NEAR PALISADE, COLORADO
COLORADO RIVER - FRUITA, COLORADO
COLORADO RIVER - ABOVE MOAB, UTAH
COLORADO RIVER - BELOW MOAB, UTAH
COLORADO RIVER - PAGE, ARIZONA
COLORADO RIVER (LAKE MEAD) - BOULDER CITY, NEVADA
COLORADO RIVER (LAKE HAVASU) - PARKER DAM, CALIFORNIA
COLORADO RIVER - YUMA, ARIZONA
pCi/l
1962
1963
1964

Figure 2. Dissolved radium–226 in Colorado River waters

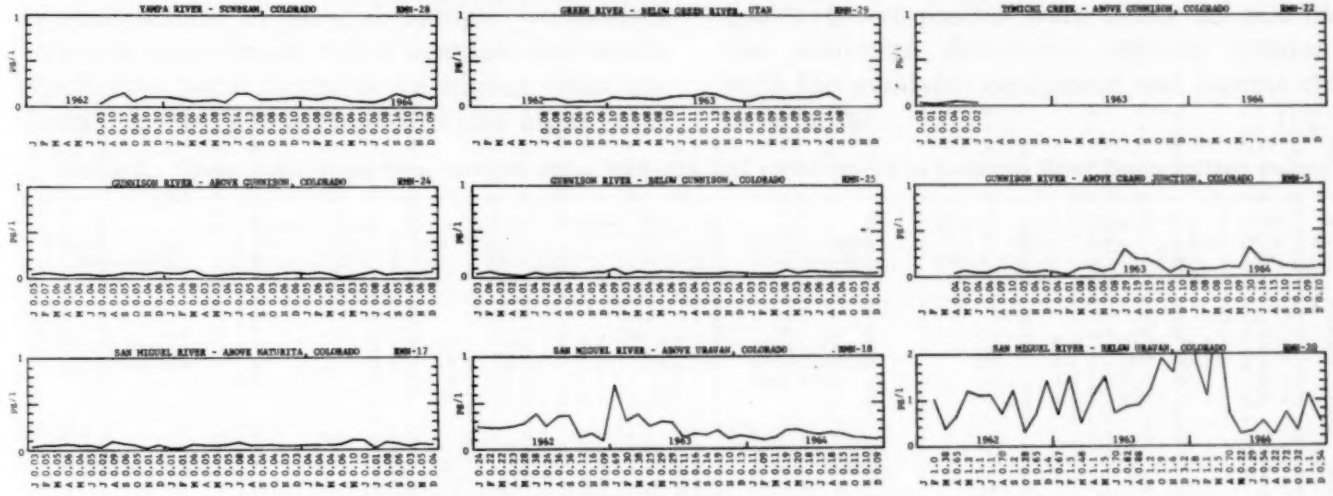
YAMPA RIVER - SUNBEAM, COLORADO
GREEN RIVER - BELOW GREEN RIVER, UTAH
TOMICHI CREEK - ABOVE GUNNISON, COLORADO
GUNNISON RIVER - ABOVE GUNNISON, COLORADO
GUNNISON RIVER - BELOW GUNNISON, COLORADO
GUNNISON RIVER - ABOVE GRAND JUNCTION, COLORADO
SAN MIGUEL RIVER - ABOVE NATURITA, COLORADO
SAN MIGUEL RIVER - ABOVE URAVAN, COLORADO
SAN MIGUEL RIVER - BELOW URAVAN, COLORADO
pCi/l
1962
1963
1964

Figure 3. Dissolved radium–226 in Yampa, Green, Tomichi, Gunnison, and San Miguel River waters

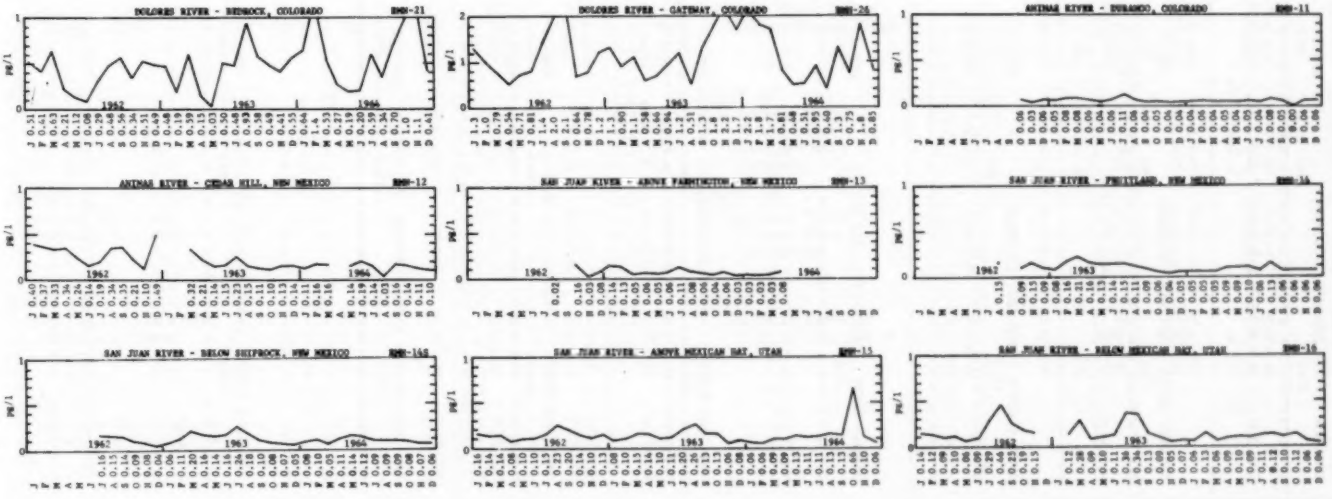
DOLORES RIVER - BEDROCK, COLORADO
DOLORES RIVER - GATEWAY, COLORADO
ANIMAS RIVER - DURANGO, COLORADO
ANIMAS RIVER - CEDAR HILL, NEW MEXICO
SAN JUAN RIVER - ABOVE FARMINGTON, NEW MEXICO
SAN JUAN RIVER - FRUITLAND, NEW MEXICO
SAN JUAN RIVER - BELOW SHIPROCK, NEW MEXICO
SAN JUAN RIVER - ABOVE MEXICAN HAT, UTAH
SAN JUAN RIVER - BELOW MEXICAN HAT, UTAH
pCi/l
1962
1963
1964

Figure 4. Dissolved radium–226 in Dolores, Animas, and San Juan River waters

Table 2. Dissolved uranium in Colorado River Basin surface waters

COLORADO RIVER - STATE LINE, COLORADO — RMN-1

1962	
O	3.3
N	3.2
D	3.6
1963	
J	3.1
F	3.0
M	3.9
A	2.6
M	0.1
J	0.8
J	2.3
A	3.9
S	2.9
O	3.0
N	3.4
D	4.4
1964	
J	3.4
F	2.7
M	3.2
A	3.5
M	1.5
J	1.2
J	2.7
A	2.2
S	2.9
O	3.5
N	3.9
D	4.3

COLORADO RIVER - DE BEQUE, COLORADO — RMN-4

1962	
O	6.5
N	4.5
D	4.2
1963	
J	4.3
F	4.4
M	5.1
A	3.2
M	0.4
J	0.7
J	2.5
A	3.3
S	3.6
O	4.2
N	4.2
D	5.5
1964	
J	4.5
F	4.5
M	4.4
A	2.9
M	1.8
J	1.3
J	2.9
A	3.2
S	3.7
O	4.6
N	7.2
D	5.8

COLORADO RIVER - FRUITA, COLORADO — RMN-6

1962	
O	8.5
N	8.5
D	10
1963	
J	9.9
F	9.5
M	8.1
A	6.2
M	4.0
J	4.7
J	12
A	12
S	14
O	14
N	11
D	13
1964	
J	10
F	9.2
M	7.6
A	10
M	5.7
J	3.2
J	8.9
A	8.8
S	13
O	14
N	12
D	14

COLORADO RIVER - ABOVE MOAB, UTAH — RMN-9

1962	
O	9.3
N	10
D	11
1963	
J	16
F	11
M	12
A	6.5
M	8.8
J	4.7
J	14
A	17
S	14
O	14
N	11
D	16
1964	
J	12
F	11
M	13
A	9.7
M	5.6
J	3.6
J	7.8
A	12
S	9.6
O	14
N	15
D	14

COLORADO RIVER - BELOW MOAB, UTAH — RMN-10

1962	
O	9.0
N	12
D	13
1963	
J	16
F	13
M	14
A	10
M	9.2
J	5.4
J	12
A	17
S	14
O	16
N	12
D	16
1964	
J	12
F	13
M	13
A	9.7
M	5.6
J	3.7
J	8.5
A	8.6
S	16
O	18
N	17
D	16

COLORADO RIVER - PAGE, ARIZONA — RMN-31

1962	
O	
N	
D	
1963	
J	
F	
M	
A	
M	13
J	11
J	9.2
A	7.4
S	7.1
O	7.9
N	8.6
D	8.6
1964	
J	8.4
F	10
M	7.5
A	11
M	9.1
J	9.2
J	11
A	10
S	6.9
O	6.5
N	7.8
D	7.8

COLORADO RIVER - BOULDER CITY, NEVADA — RMN-32

1962	
O	
N	
D	
1963	
J	
F	
M	10
A	8.8
M	8.0
J	7.8
J	7.4
A	7.0
S	7.9
O	8.6
N	5.6
D	8.1
1964	
J	7.4
F	8.6
M	9.0
A	9.6
M	7.9
J	6.9
J	8.9
A	9.2
S	8.8
O	11
N	8.8
D	11

COLORADO RIVER - PARKER DAM, CALIFORNIA — RMN-33

1962	
O	
N	
D	
1963	
J	9.9
F	11
M	11
A	9.9
M	8.2
J	8.1
J	8.8
A	6.6
S	7.2
O	6.2
N	5.9
D	7.4
1964	
J	7.7
F	8.7
M	7.9
A	8.1
M	8.8
J	7.2
J	7.9
A	8.1
S	9.4
O	8.6
N	8.9
D	8.5

COLORADO RIVER - YUMA, ARIZONA — RMN-30

1962	
O	9.6
N	8.3
D	8.0
1963	
J	9.5
F	11
M	8.8
A	9.2
M	9.6
J	9.2
J	7.2
A	7.9
S	7.6
O	6.7
N	5.9
D	7.1
1964	
J	7.1
F	7.3
M	9.0
A	8.2
M	7.9
J	7.5
J	8.8
A	8.1
S	8.1
O	9.2
N	9.2
D	8.8

YAMPA RIVER - SUNBEAM, COLORADO — RMN-28

1962	
O	2.4
N	2.3
D	2.0
1963	
J	2.8
F	4.9
M	8.0
A	1.5
M	1.8
J	1.2
J	4.0
A	3.1
S	1.8
O	2.5
N	0.6
D	2.8
1964	
J	0.8
F	1.9
M	0.9
A	7.9
M	1.8
J	0.6
J	1.7
A	2.4
S	3.5
O	3.5
N	2.9
D	3.6

GREEN RIVER - BELOW GREEN RIVER, UTAH — RMN-29

1962	
O	6.0
N	7.3
D	6.2
1963	
J	6.0
F	5.1
M	7.6
A	3.2
M	1.6
J	2.6
J	4.8
A	5.5
S	6.1
O	7.4
N	5.4
D	5.0
1964	
J	5.0
F	4.1
M	3.9
A	6.3
M	3.6
J	3.3
J	3.5
A	5.5
S	5.2
O	
N	
D	

TOMICHI CREEK - ABOVE GUNNISON, COLORADO — RMN-22

1962	
O	
N	
D	
1963	
J	
F	
M	
A	
M	
J	
J	
A	
S	
O	
N	
D	
1964	
J	
F	
M	
A	
M	
J	
J	
A	
S	
O	
N	
D	

GUNNISON RIVER - ABOVE GUNNISON, COLORADO — RMN-24

1962	
O	1.3
N	1.2
D	1.9
1963	
J	2.6
F	1.2
M	0.7
A	0.5
M	0.9
J	0.6
J	1.0
A	0.0
S	0.6
O	1.1
N	0.1
D	2.5
1964	
J	1.3
F	1.3
M	0.7
A	1.5
M	1.5
J	0.6
J	1.0
A	1.4
S	1.2
O	1.7
N	1.3
D	1.9

GUNNISON RIVER - BELOW GUNNISON, COLORADO — RMN-25

1962	
O	3.0
N	2.3
D	2.6
1963	
J	3.2
F	3.2
M	2.0
A	0.9
M	0.4
J	1.0
J	0.6
A	0.7
S	1.5
O	1.6
N	1.3
D	3.6
1964	
J	2.7
F	2.9
M	1.8
A	2.9
M	2.2
J	1.0
J	1.9
A	1.6
S	2.0
O	2.4
N	4.3
D	4.3

GUNNISON RIVER - ABOVE GRAND JUNCTION, COLORADO — RMN-5

1962	
O	12
N	13
D	13
1963	
J	13
F	10
M	10
A	4.7
M	2.9
J	0.7
J	12
A	14
S	17
O	15
N	13
D	14
1964	
J	12
F	11
M	12
A	4.4
M	3.5
J	4.0
J	8.9
A	10
S	14
O	19
N	17
D	14

SAN MIGUEL RIVER - ABOVE NATURITA, COLORADO — RMN-17

1962	
O	1.2
N	1.0
D	0.9
1963	
J	1.1
F	1.3
M	2.1
A	0.1
M	0.0
J	0.5
J	2.3
A	0.8
S	1.6
O	3.9
N	0.6
D	2.0
1964	
J	1.2
F	1.0
M	0.1
A	1.5
M	0.9
J	0.0
J	1.0
A	1.2
S	2.7
O	2.3
N	2.4
D	2.2

SAN MIGUEL RIVER - ABOVE URAVAN, COLORADO — RMN-18

1962	
O	5.0
N	6.5
D	7.0
1963	
J	31
F	11
M	8.9
A	3.2
M	2.2
J	2.1
J	7.7
A	3.8
S	3.1
O	6.0
N	1.9
D	5.7
1964	
J	3.2
F	3.9
M	2.5
A	3.2
M	1.8
J	0.9
J	3.7
A	3.5
S	6.3
O	3.5
N	5.0
D	5.0

SAN MIGUEL RIVER - BELOW URAVAN, COLORADO — RMN-20

1962	
O	11
N	21
D	57
1963	
J	60
F	27
M	27
A	16
M	10
J	1.8
J	17
A	44
S	38
O	31
N	16
D	22
1964	
J	28
F	55
M	49
A	52
M	2.3
J	5.0
J	19
A	20
S	41
O	35
N	59
D	44

DOLORES RIVER - BEDROCK, COLORADO — RMN-21

1962	
O	11
N	12
D	11
1963	
J	8.9
F	7.4
M	9.8
A	2.9
M	0.9
J	6.3
J	11
A	31
S	28
O	35
N	14
D	19
1964	
J	16
F	16
M	17
A	6.8
M	2.6
J	2.0
J	11
A	8.5
S	21
O	29
N	30
D	12

DOLORES RIVER - GATEWAY, COLORADO — RMN-26

1962	
O	11
N	3.0
D	3.3
1963	
J	12
F	14
M	1.9
A	9.5
M	4.3
J	3.1
J	15
A	25
S	29
O	16
N	22
D	35
1964	
J	28
F	
M	51
A	34
M	6.8
J	4.0
J	13
A	9.2
S	33
O	31
N	50
D	33

ANIMAS RIVER - DURANGO, COLORADO — RMN-11

1962	
O	1.0
N	1.8
D	0.6
1963	
J	1.2
F	2.1
M	1.2
A	0.2
M	2.5
J	0.9
J	1.5
A	0.5
S	1.0
O	0.8
N	1.1
D	1.5
1964	
J	1.0
F	1.1
M	0.4
A	0.9
M	0.6
J	0.7
J	0.7
A	0.9
S	1.4
O	1.9
N	1.9
D	1.9

ANIMAS RIVER - CEDAR HILL, NEW MEXICO — RMN-12

1962	
O	7.4
N	8.7
D	6.0
1963	
J	
F	
M	6.9
A	2.0
M	2.0
J	1.2
J	2.3
A	2.2
S	1.8
O	4.4
N	4.0
D	4.4
1964	
J	2.4
F	2.5
M	2.5
A	1.3
M	0.7
J	1.4
J	2.3
A	3.7
S	6.0
O	3.7
N	3.5
D	

SAN JUAN RIVER - ABOVE FARMINGTON, NEW MEXICO — RMN-13

1962	
O	8.3
N	5.4
D	4.3
1963	
J	4.0
F	5.5
M	1.9
A	0.8
M	3.6
J	1.9
J	1.7
A	4.5
S	2.3
O	1.1
N	0.0
D	2.6
1964	
J	1.2
F	1.3
M	1.2
A	2.2
M	
J	
J	
A	
S	
O	
N	
D	

SAN JUAN RIVER - FRUITLAND, NEW MEXICO — RMN-14

1962	
O	5.1
N	7.0
D	6.8
1963	
J	6.6
F	7.7
M	9.0
A	1.9
M	3.2
J	2.8
J	2.7
A	4.2
S	2.6
O	2.9
N	2.2
D	3.6
1964	
J	2.5
F	3.0
M	2.0
A	2.4
M	2.1
J	0.9
J	1.6
A	2.3
S	3.5
O	3.2
N	4.0
D	4.0

SAN JUAN RIVER - BELOW SHIPROCK, NEW MEXICO — RMN-14S

1962	
O	8.3
N	11
D	13
1963	
J	12
F	16
M	14
A	2.3
M	11
J	7.5
J	22
A	13
S	5.4
O	18
N	16
D	14
1964	
J	15
F	3.4
M	2.7
A	31
M	36
J	6.9
J	6.5
A	5.4
S	11
O	18
N	16
D	14

SAN JUAN RIVER - ABOVE MEXICAN HAT, UTAH — RMN-15

1962	
O	8.7
N	9.0
D	9.8
1963	
J	13
F	8.9
M	11
A	5.7
M	5.9
J	5.7
J	11
A	12
S	6.3
O	7.6
N	4.5
D	6.8
1964	
J	5.5
F	6.2
M	5.9
A	7.9
M	5.9
J	2.6
J	3.0
A	5.8
S	9.5
O	23
N	8.8
D	7.8

SAN JUAN RIVER - BELOW MEXICAN HAT, UTAH — RMN-16

1962	
O	10
N	11
D	9.0
1963	
J	0.2
F	15
M	12
A	14
M	6.2
J	6.0
J	8.4
A	10
S	12
O	11
N	6.7
D	7.3
1964	
J	7.5
F	12
M	8.8
A	8.5
M	8.9
J	3.4
J	3.3
A	5.8
S	16
O	24
N	19
D	14

above the currently accepted yearly average limit of 3 pCi/liter for radium in drinking water (*7*). No network sampling station has exhibited an overall average above this limit and only two stations have sustained average radium–226 concentrations at or slightly above the 1 pg/liter suggested maximum desirable level for Colorado River Basin waters (*8*).

The quarterly gross alpha figures given in table 3 agree with the general range of monthly figures reported by the Public Health Service Water Pollution Surveillance System (WPSS) for their Colorado River Basin stations, while the gross beta results tend to range somewhat lower than the WPSS results.

The lead–210 and thorium alpha results are consistently low throughout the Basin, but concentrations in several sample composites collected immediately below uranium mill waste discharges were found to be higher than upstream levels. The highest thorium alpha level of 8.0 pCi/liter reported for the second quarter below Uravan, Colorado, may be erroneous in view of the lower levels obtained for the other three quarters.

Four of the five river sampling stations, which are subject to flow fluctuations and for which strontium–90 determinations were performed, tend to show higher-than-average strontium–90 levels during July–September 1963. Additionally, several slightly higher levels occurred in the April–June and July–September 1964 quarters, suggesting a seasonal variation related to runoff collection of fallout. The other two stations which are sampled for strontium–90 are in regulated flow "reaches" and are not immediately affected by runoff.

All gross gamma determinations on the quarterly composites were below 33 pCi/liter, the minimum detectable activity obtainable with the available equipment and sample characteristics.

Table 3. Gross alpha, gross beta, thorium alpha, lead–210, and strontium–90 in Colorado River Basin surface waters

Station number and location	Quarter[1] period	Gross alpha (pCi/liter)	Gross beta (pCi/liter)	Th-alpha (pCi/liter)	^{210}Pb (pCi/liter)	^{90}Sr (pCi/liter)
RMN 1						
Colorado River at	1	2	10	NA	NA	NA
Silt, Colorado	2	2	18	NA	0.4	NA
	3	2	8	0.4	0.1	NA
	4	3	20	0.1	0.1	NA
RMN 4						
Colorado River at	1	3	11	NA	NA	2.8
DeBeque, Colorado	2	3	22	NA	NA	0.9
	3	3	9	NA	NA	0.9
	4	1	23	NA	NA	1.8
RMN 6						
Colorado River below	1	7	41	0.1	0.1	NA
Grand Junction,	2	5	11	0.0	0.3	NA
Colorado	3	7	22	0.5	0.1	NA
	4	2	14	0.1	0.0	NA
RMN 9						
Colorado River above	1	8	33	NA	NA	8.4
Moab, Utah	2	12	20	NA	NA	1.6
	3	5	12	NA	NA	1.2
	4	2	16	NA	NA	2.2
RMN 10						
Colorado River below	1	9	31	0.1	0.0	NA
Moab, Utah	2	4	8	0.2	0.1	NA
	3	6	16	0.0	0.1	NA
	4	3	14	0.0	0.0	NA
RMN 11						
Animas River at	1	1	5	NA	NA	0.9
Durango, Colorado	2	1	5	NA	NA	0.7
	3	1	9	NA	NA	0.9
	4	1	8	NA	NA	1.5
RMN 14S						
San Juan River below	1	12	16	0.0	0.1	NA
Shiprock, New	2	7	14	0.1	0.0	NA
Mexico	3	3	9	0.0	0.0	NA
	4	8	14	0.2	0.0	NA
RMN 15						
San Juan River above	1	6	29	NA	NA	6.0
Mexican Hat, Utah	2	4	14	NA	NA	3.2
	3	4	10	NA	NA	2.0
	4	3	14	NA	NA	2.9
RMN 16						
San Juan River	1	8	30	0.4	0.2	NA
below Mexican	2	5	13	0.3	0.1	NA
Hat, Utah	3	4	30	0.2	0.2	NA
	4	4	10	0.3	0.2	NA
RMN 17						
San Miguel River	1	2	7	0.1	0.0	NA
above Naturita,	2	2	7	0.1	0.0	NA
Colorado	3	2	7	0.1	0.1	NA
	4	1	10	0.0	0.0	NA
RMN 20						
San Miguel River	1	13	23	1.1	0.6	NA
below Uravan,	2	19	43	8.0	0.4	NA
Colorado	3	20	26	0.8	0.2	NA
	4	12	33	1.0	0.1	NA
RMN 29						
Green River below	1	3	26	NA	NA	9.6
Green River, Utah	2	3	14	NA	NA	3.3
	3	1	10	NA	NA	2.5
	4	3	12	NA	NA	3.2
RMN 32						
Colorado River	1	3	5	NA	NA	1.1
(Lake Mead) at	2	3	10	NA	NA	1.2
Boulder City,	3	4	4	NA	NA	1.2
Nevada	4	5	9	NA	NA	1.4
RMN 33						
Colorado River at	1	5	9	0.0	0.2	NA
Parker Dam,	2	3	22	0.0	0.2	NA
California	3	4	9	0.1	0.1	NA
	4	4	11	0.2	0.0	NA
RMN 30						
Colorado River at	1	6	8	NA	NA	1.2
Yuma, Arizona	2	4	10	NA	NA	1.5
	3	3	11	NA	NA	1.3
	4	4	10	NA	NA	1.5

[1] Quarter period
1 July—September 1963
2 October—December 1963
3 January—March 1964
4 April—June 1964
NA indicates no analysis performed.

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(*8*) DIVISION OF WATER SUPPLY AND POLLUTION CONTROL, PUBLIC HEALTH SERVICE. Transcript, Third Session Conference of Interstate Waters of the Colorado River and its Tributaries. U.S. Department of Health, Education, and Welfare, Region VIII, Denver, Colorado (May 1962).

RADIOACTIVITY IN FLORIDA WATERS, 1964[1]

Division of Radiological and Occupational Health, Florida State Board of Health

The Florida State Board of Health samples raw surface water from locations in 13 hydrological subbasins. In nine of these subbasins, raw ground water is also sampled. The samples are collected on a variable frequency and analyzed for gross alpha and beta radioactivity.

Figure 1 shows the locations of the surface water subbasins from which samples are obtained. Average gross alpha and gross beta radioactivity levels in raw surface waters during 1964 are presented in table 1, while table 2 gives the radioactivity levels in ground waters.

The average gross beta radioactivity in water supplies is well below the standard for gross beta radioactivity in drinking water (1,000 pCi/liter), as is average gross alpha radioactivity when compared to its standard for drinking water (3 pCi/liter when radium is present) (*1*).

In addition to the sampling being done by the Florida State Board of Health, the Sanitary Engineering Research Laboratory, University of Florida, samples raw surface water and water supplies in Alachua County and analyzes the samples for gross alpha and gross beta radioactivity. Samples are taken monthly except at Gainesville, where raw water is sampled daily. These data are summarized in table 1 as quarterly and yearly averages.

The yearly average beta activity in surface waters was more than double that in ground waters. This additional beta activity in surface waters is attributed to fallout. The alpha activity, which is attributed to natural activity of minerals, is very nearly the same for ground and surface water. The presence of gross beta activity due to fallout is clearly evident when comparing gross beta radioactivity

[1] Data taken from "Report of Florida Radiological Data, Environmental Monitoring Programs, 1964," Florida State Board of Health, Bureau of Preventable Diseases, Division of Radiological and Occupational Health, Jacksonville, Florida.

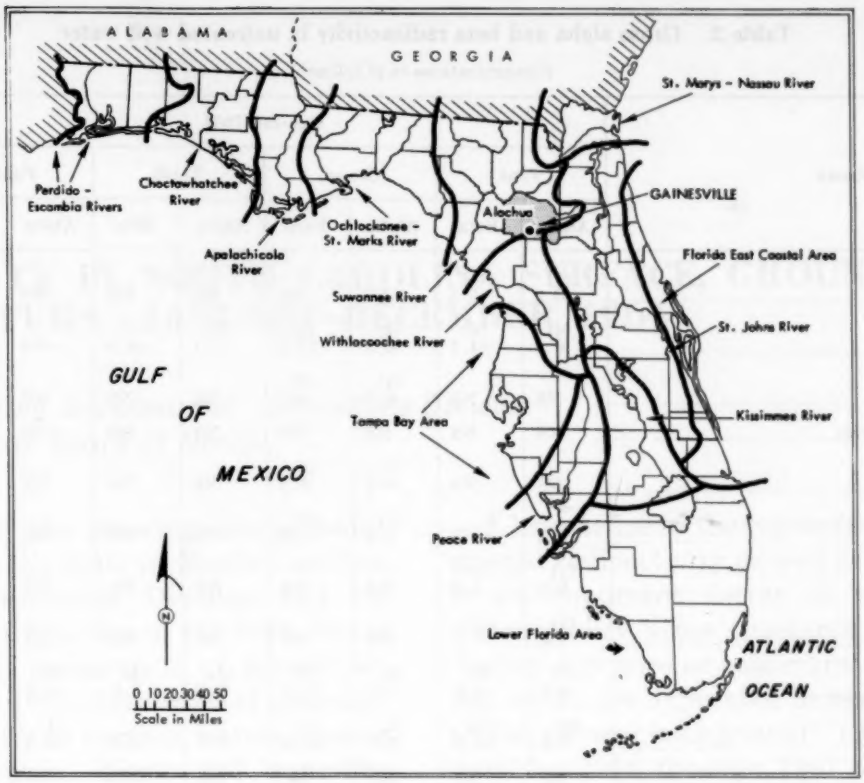

Figure 1. Hydrological surface subbasins in Florida

Table 1. Gross alpha and beta radioactivity in raw surface water, 1965

(Concentrations in pCi/liter)

Subbasin	Quarter, 1964								Yearly average	
	First		Second		Third		Fourth			
	Alpha	Beta	Alpha	Beta	Alpha	Beta	Alpha	Beta	Alpha	Beta
St. Marys—Nassau River	(1) 0.2	(1) 61.5	NS	NS	NS	NS	(2) 0.2	(2) 24.0	(3) 0.2	(3) 36.5
St. Johns River	(25) 0.3	(25) 60.1	(17) 0.2	(17) 55.2	(17) 0.6	(17) 79.2	(15) 0.4	(15) 71.1	(74) 0.4	(74) 65.6
Suwannee River	(2) 0.1	(2) 74.2	(10) 0.4	(10) 32.2	NS	NS	NS	NS	(12) 0.4	(12) 39.2
Ochlockonee—St. Marys River	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS
Withlacoochee River	NS	NS	(7) 0.1	(7) 32.1	NS	NS	(3) 0.2	(3) 38.2	(10) 0.1	(10) 33.9
Tampa Bay Area	(2) 0.0	(2) 38.5	(3) 0.4	(3) 18.0	NS	NS	NS	NS	(5) 0.2	(5) 26.2
Peace River	(3) 0.7	(3) 57.9	(2) 0.6	(2) 59.5	NS	NS	NS	NS	(5) 0.7	(5) 58.5
Kissimmee River	(5) 0.4	(5) 55.8	(2) 0.2	(2) 42.4	(1) 0.4	(1) 46.2	(2) 0.4	(2) 31.2	(10) 0.4	(10) 47.2
Florida East Coastal Area	(21) 0.3	(21) 58.9	(7) 0.3	(7) 78.2	(13) 0.4	(13) 139.0	NS	NS	(41) 0.3	(41) 87.6
Lower Florida Area	(5) 0.4	(5) 50.9	(2) 0.4	(2) 60.7	NS	NS	(2) 0.2	(2) 41.7	(9) 0.4	(9) 51.0
Apalachicola River	(4) 0.3	(4) 20.0	NS	NS	NS	NS	(1) 0.4	(1) 16.6	(5) 0.3	(5) 19.3
Choctawhatchee	(6) 0.3	(6) 17.2	NS	NS	NS	NS	(2) 0.1	(2) 8.0	(8) 0.3	(8) 14.9
Perdido—Escambia Rivers	(4) 0.3	(4) 32.3	NS	NS	NS	NS	(2) 0.7	(2) 11.3	(6) 0.4	(6) 25.3
Total samples	(78)	(78)	(50)	(50)	(31)	(31)	(29)	(29)	(188)	(188)
Average	0.3	52.0	0.3	48.2	0.5	103.2	0.3	49.3	0.3	59.0

Note: Number in parentheses indicates number of samples.
NS indicates no sample collected.

Table 2. Gross alpha and beta radioactivity in untreated well water

(Concentrations in pCi/liter)

Subbasin	Quarter, 1964								Yearly average	
	First		Second		Third		Fourth			
	Alpha	Beta	Alpha	Beta	Alpha	Beta	Alpha	Beta	Alpha	Beta
St. Marys-Nassau River	(1) 0.2	(1) 5.5	NS	NS	NS	NS	NS	NS	(1) 0.2	(1) 5.5
St. Johns River	(4) 0.2	(4) 34.1	(2) 0.3	(2) 32.7	(3) 0.1	(3) 16.3	NS	NS	(9) 0.2	(9) 27.9
Suwannee River	NS	NS	(2) 0.2	(2) 4.5	NS	NS	NS	NS	(2) 0.2	(2) 4.5
Ochlockonee—St. Marys River	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS
Withlacoochee River	NS	NS	(2) 0.6	(2) 3.5	NS	NS	NS	NS	(2) 0.6	(2) 3.5
Tampa Bay Area	NS	NS	(1) 0.3	(1) 5.0	NS	NS	NS	NS	(1) 0.3	(1) 5.0
Peace River	(1) 0.6	(1) 4.9	NS	NS	NS	NS	NS	NS	(1) 0.6	(1) 4.9
Kissimmee River	(2) 0.3	(2) 7.1	NS	NS	NS	NS	NS	NS	(2) 0.3	(2) 7.1
Florida East Coastal Area	(7) 0.2	(7) 8.8	(2) 0.2	(2) 43.3	(4) 0.3	(4) 8.6	(3) 0.2	(3) 12.8	(16) 0.2	(16) 13.8
Lower Florida Area	(5) 0.8	(5) 58.8	(4) 0.1	(4) 50.9	NS	NS	(1) 0.8	(1) 5.5	(10) 0.5	(10) 50.3
Apalachicola River	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS
Choctawhatchee River	NS	NS	NS	NS	NS	NS	NS	NS	NS	NS
Perdido—Escambia Rivers	(1) 0.8	(1) 10.3	NS	NS	NS	NS	NS	NS	(1) 0.8	(1) 10.3
Total samples	(21)	(21)	(13)	(13)	(7)	(7)	(4)	(4)	(45)	(45)
Average	0.4	26.1	0.3	28.9	0.2	11.9	0.3	11.0	0.3	23.4

Note: Number in parentheses indicates number of samples.
NS indicates no sample collected.

in raw surface water and raw well water. However, no obvious trend in gross beta radioactivity levels as a function of fallout was indicated by the data obtained.

REFERENCE

(1) PUBLIC HEALTH SERVICE. Public Health Service drinking water standards, revised 1962. Publication No. 956. U.S. Government Printing Office, Washington, D.C. 20402 (1963). Price 30 cents.

RADIOACTIVITY IN NORTH CAROLINA SURFACE, GROUND, AND CISTERN WATERS,[1] JANUARY–DECEMBER 1964

Sanitary Engineering Division and Laboratory Division
North Carolina State Board of Health

In recognition of the growing use of radioactive materials in the State of North Carolina, the Sanitary Engineering Division and the State Laboratory of Hygiene of the State Board of Health, with the cooperation of municipalities, began in June 1958 a program of measurement of radioactivity in surface waters used as public water supplies. Some 147 sampling points were established to sample raw surface water. In July 1962, the program was expanded to include the sampling of cistern water from Ocracoke Island. Then, in January 1963, other types of environmental samples were also collected (*1*), two of which were treated water from surface water supplies and ground water from wells used as municipal water supplies.

All waters contain traces of radioactivity originating from naturally radioactive minerals dissolved from rock strata or from radioactive particulate material or gases in the atmosphere. Common among these materials are trace elements of potassium–40, radium, thorium, and uranium. Such trace elements are dissolved by water both on its way to and flowing in the watercourses. Precipitation is the major mechanism by which particulate matter or radioactive gases, such as thoron and radon, are removed from the atmosphere. The combined radioactivity of these materials constitutes what is known as "background radioactivity" of the water. The total activity would include both background radioactivity and contributions from fallout and other man-made sources.

A knowledge of the concentration of the background radioactivity as well as the total activity is an important factor in the appraisal of water quality, since standards pertaining to radiation exposure or concentration within drinking water are expressed in terms of "additions to the natural background" (*2*). Surface water data for 1958 through 1961 are considered as background radioactivity and have previously been published in *Radiological Health Data* (*3*).

Laboratory procedure

Total solids content is determined for each sample so that proper sample proportions may be used in analysis. The calculated amount of a sample is concentrated to a few milliliters on a steam bath. The residue is transferred to a stainless-steel planchet and evaporated to dryness under an infrared light. After further drying for 1 hour at 103° C., the sample is cooled in a desiccator and is then counted.

Alpha-plus-beta counting is done in a gas-flow internal proportional counter calibrated with a radium D+E source. Taking instrument background (17 counts per minute) into consideration, each sample is counted for a preset count and the results are reported in terms of pCi/liter.

Results

Table 1 gives the average gross (alpha-plus-beta) radioactivity in raw surface water as well as the maximum and minimum values recorded during 1964. The first column consists of code numbers which indicate the geographical positions of the sampling stations, as shown in figure 1.

[1] Summarized from "Background Radioactivity in Surface Water Supplies of North Carolina, 1958–1962" and "Environmental Surveillance, 1964."

Table 1. Gross radioactivity found in surface waters used as sources of public water supplies in North Carolina, 1964

(Radioactivity concentrations in pCi/liter)

Code	Location	Source of water supply	Raw Water				Treated Water			
			No. of samples	Average	Maximum	Minimum	No. of samples	Average	Maximum	Minimum
1	Alamance	Big Alamance Creek	1	12.2			0			
2	Albemarle	Yadkin River	11	9.3	16.4	6.8	7	5.5	9.6	3.2
3	Andrews	Beaver Creek	2	3.7	3.9	3.6	0			
3a	Apex		2	19.6	25.2	13.1	0			
4	Asheboro	Back Creek	3	18.7	30.6	12.0	1	7.5		
5	Asheville	Bee Tree Creek	11	4.5	10.8	2.0	7	5.7	12.3	2.4
6	Badin	Yadkin River	2	11.8	15.1	8.5	0			
7	Belmont	Catawba River	4	7.1	17.4	1.0	0			
7a	Bessemer City		1	7.4			0			
8	Biscoe	Little River	4	20.3	43.6	6.7	0			
9	Blowing Rock	Brickhouse Creek	3	8.8	14.0	3.7	0			
9a	Boone		2	4.0	4.1	3.9	0			
9b	Brevard		2	3.0	3.6	2.4	0			
10	Bryson City	Lands Creek	1	3.3			0			
10a	Burgaw		2	23.4	25.8	21.7	0			
11	Burlington	Stony Creek	12	14.5	26.7	6.0	7	9.9	17.2	6.3
11a	Candor		3	15.9	25.8	5.8	0			
12	Canton	Pigeon River; Ruff Creek	2	3.1	4.2	2.1	0			
13	Carthage	Spring	2	8.1	8.8	7.5	0			
14	Chapel Hill	Price Creek	2	12.4	13.0	11.9	0			
15	Charlotte	Catawba River	11	8.6	16.6	5.1	8	5.7	9.6	2.3
16	Cliffside	Second Broad River	3	7.4	11.3	5.3	0			
17	Clinton	Black River	2	10.2	13.1	7.3	0			
18	Concord	Cold Water Creek & Lumby Creek	10	17.9	36.2	11.6	8	9.0	13.8	3.8
19	Cooleemee	South Fork Yadkin	2	7.7	8.3	7.3	0			
20	Cramerton	South Fork Catawba River	10	7.3	13.9	3.4	8	6.6	16.2	3.2
21	Creedmoor	Ledge Creek	2	14.9	16.5	13.3	0			
22	Cullowhee	Long Branch; Flat Creek	2	1.9	2.3	1.4	0			
23	Davidson	Cathy's Creek	2	8.0	10.5	5.7	0			
24	Denton	Lick Branch	2	15.1	19.5	10.0	0			
25	Dunn	Cape Fear River	3	15.7	22.9	11.2	0			
26	Durham	Flat River	11	14.8	21.1	9.7	8	8.3	17.1	2.2
27	Elizabeth City	Pasquotank River	0				1	10.0		
29	Elkin	Big Elkin Creek	2	5.8	9.0	2.6	0			
30	Fayetteville	Cape Fear River	11	18.4	38.0	5.9	8	9.3	18.0	3.3
31	Forest City	Second Broad River	2	7.1	9.0	5.2	0			
32	Fontana Dam	Little Tennessee River	1	5.0			0			
33	Franklinton	Kearney's Creek	1	6.2			0			
34	Gastonia	Long Creek; Catawba River	11	8.0	15.7	4.6	7	6.3	8.3	5.2
35	Goldsboro	Little River; Neuse River	2	18.1	20.2	16.1	1	7.4		
36	Graham	Back Creek	3	26.9	64.7	5.3	0			
37	Granite Falls	Catawba River	1	3.9			0			
38	Greensboro	Reedy Fork Creek	11	13.6	27.7	10.2	8	9.2	17.3	2.2
39	Greenville	Tar River	11	14.8	29.3	7.1	7	9.6	24.4	4.0
40	Hamlet	Marks Creek	1	7.0			0			
41	Henderson	Sandy Creek	0				0			
42	Hendersonville	North Fork Mills River and Bradley Creek	1	0.5			0			
43	Hertford	Perquimans River	0				0			
44	Hickory	Catawba River	10	7.8	16.2	4.2	8	6.7	15.2	4.4
45	High Point	Deep River	10	15.3	22.9	9.4	7	9.3	20.0	8.4
46	Hillsboro	Eno River	2	5.4	8.5	2.6	0			
47	Jamestown	Deep River	0				0			
48	Kannapolis	Buffalo Creek	12	15.2	33.4	8.5	7	8.7	11.6	3.8
49	Kernersville	Belew's Creek	1	9.0			0			
50	Kings Mountain	Kings Creek	1	4.6			0			
51	Landis	Grant's Creek	1	10.6			0			
52	Laurinburg	Jordan Creek	11	9.2	17.5	3.3	7	7.4	12.0	3.3
53	Leaksville	Dan River	3	5.4	7.8	4.3	0			
54	Lenoir	Zacks Fork Creek & Catawba River	3	9.8	16.9	4.6	0			
55	Lexington	Abbott's Creek	3	25.8	52.3	12.2	0			
56	Lillington	Cape Fear River	1	11.0			0			
57	Lincolnton	Walker Branch	2	6.3	7.8	4.9	0			
58	Louisburg	Tar River	1	3.5			0			
59	Lumberton	Lumber River	10	16.5	14.3	6.2	7	9.1	14.3	6.2
60	Marion	Mackey's Creek; Buck Creek	1	2.6			0			
60a	Mars Hill		1	1.8			0			
60b	Marshall		1	3.6			0			
61	Mayodan	Mayo River	1	17.4			0			
62	Mebane	Mills Creek	1	7.1			0			
63	Mocksville	Bear Creek	1	5.4			0			
64	Monroe	Richardson Creek	2	23.5	28.7	17.5	0			
65	Mooresville	Byers Creek; Catawba River	3	18.7	28.3	9.4	0			
66	Morganton	Henry River	3	2.6	3.1	2.2	0			
67	Mount Airy	Lovell's Creek	3	5.2	7.4	3.7	0			
68	Mount Gilead	Pee Dee River	3	12.7	17.2	9.7	0			
69	Mount Holly	Catawba River	10	9.5	16.7	3.4	6	7.3	11.5	5.5
70	Mount Pleasant	Dutch Buffalo Creek	1	13.7			0			

Table 1. Gross radioactivity found in surface waters used as sources of public water supplies in North Carolina, 1964 —Continued

(Radioactivity concentrations in pCi/liter)

Code	Location	Source of water supply	Raw Water				Treated Water			
			No. of samples	Average	Maximum	Minimum	No. of samples	Average	Maximum	Minimum
70a	Murphy		3	5.3	8.9	2.4	0			
71	New Bern	Neuse River	0				0			
72	Newton	Jacobs Fork	1	13.5			0			
73	North Wilkesboro	Reddies River	3	8.8	16.4	2.3	0			
73a	Norwood		1	7.1			0			
74	Old Fort	Jarrett Creek	1	9.5			0			
75	Oxford	Tar River; Hatcher's Run	1	23.1			0			
75a	Pinehurst		1	2.1			0			
75b	Pilot Mountain		1	12.4			0			
76	Pisgah Forest	Davidson River	1	3.5			0			
77	Pittsboro	Robinson Creek	1	27.6			0			
78	Raleigh	Lake Raleigh; Lake Wheeler	12	16.2	33.6	5.5	7	11.4	26.8	6.7
79	Ramseur	Sandy Creek	1	35.8			0			
80	Randleman	Pole Cat Creek	1	25.4			0			
80a	Reidsville		3	10.2	12.3	8.5	0			
81	Ridgecrest	Mountain Stream	1	2.9			0			
82	Riverbend	Catawba River	1	8.1			0			
83	Roanoke Rapids	Roanoke River	3	13.2	21.7	7.0	0			
83a	Robbins		1	20.7			0			
84	Robbinsville	Rock & Burgin's Creek	1	9.7			0			
85	Rockingham	Falling Creek	2	9.4	12.7	6.2	0			
86	Rocky Mount	Tar River	10	13.1	22.3	7.0	7	7.1	9.5	3.6
87	Roxboro	Lake Issac Walton; Storey's Creek	1	16.4			0			
88	Salisbury	Yadkin River	10	13.8	49.8	5.2	7	7.0	10.5	3.1
89	Saluda	Paces Creek	0				0			
90	Sanford	Lick Creek; Potterage Creek	2	12.5	13.9	11.4	0			
91	Shelby	First Broad River	4	4.7	8.6	2.6	0			
92	Siler City	Rocky River	3	28.1	44.6	15.6	0			
93	Smithfield	Neuse River	10	20.5	36.1	7.2	8	13.4	44.9	1.9
93a	Southern Pines		1	18.9			0			
94	Spindale-Rutherfordton	Hollands Creek; Cathy's Creek	0				0			
95	Spray	Smith River	11	7.5	14.3	3.9	7	7.8	21.0	3.8
96	Spruce Pine	Beaver Creek	1	1.8			0			
97	Stanley	Hoyle's Creek	1	4.5			0			
98	Statesville	Fourth Creek	2	4.5	7.0	2.2	0			
99	Statesville	South Yadkin River	2	8.2	8.9	7.7	0			
100	Sylva	Dill's Creek	3	1.8	2.3	1.3	0			
101	Tapoco	Yellow Hammer Creek	1	2.0			0			
102	Tarboro	Tar River	3	15.9	19.2	11.8	0			
103	Thomasville	Abbott's Creek	3	11.7	23.2	1.9	0			
104	Troy	Denson Creek; Downing Creek	1	6.0			0			
105	Tryon	Falls Creek; Vaughn's Creek	1	3.9			0			
105a	Vass		1	13.9			0			
106	Valdese	Micol Creek; Hoyle Creek; Catawba River	2	9.7	12.3	7.4	0			
107	Wadesboro	Jones Creek	1	32.9			0			
108	Wake Forest	Smith's Creek	0				1	6.5		
109	Washington	Tranters Creek	1	31.6			0			
110	Waterville	Big Creek	1	2.2			0			
111	Waynesville	Cherry Cive & Shiney Creek	1	2.1			0			
112	Weaverville	Wagner Branch & Ox Creek	1	2.9			0			
113	Weldon	Roanoke River	1	12.2			0			
114	Wilkesboro	Cub Creek	1	4.3			0			
115	Wilmington	Cape Fear River	11	19.3	48.7	7.6	7	7.9	15.0	1.5
116	Wilson	Contentnea Creek	0				0			
117	Winston-Salem	Salem Creek	4	10.4	15.2	5.8	5	5.7	7.4	0.9
118	Winston-Salem	Yadkin River	7	7.8	18.8	3.7	2	3.4	4.0	2.8
119	Winton	Chowan River	1	18.9			0			
119a	Yanceyville		11	10.7			0			
120	Zebulon	Little River	3	8.8	10.5	7.3	0			

After purification of these surface waters, the finished water is distributed for domestic and industrial uses. Finished water from some of these supplies has been sampled for gross radioactivity. Table 1 gives the average, maximum, and minimum radioactivity in finished water from these supplies during 1964.

On some of the islands situated along the eastern seaboard of North Carolina (figure 2), and also at some private residences in Durham County, drinking water is obtained from small cistern water supplies. The cistern supplies are characteristically constructed with roof prewash valves which permit the diversion of

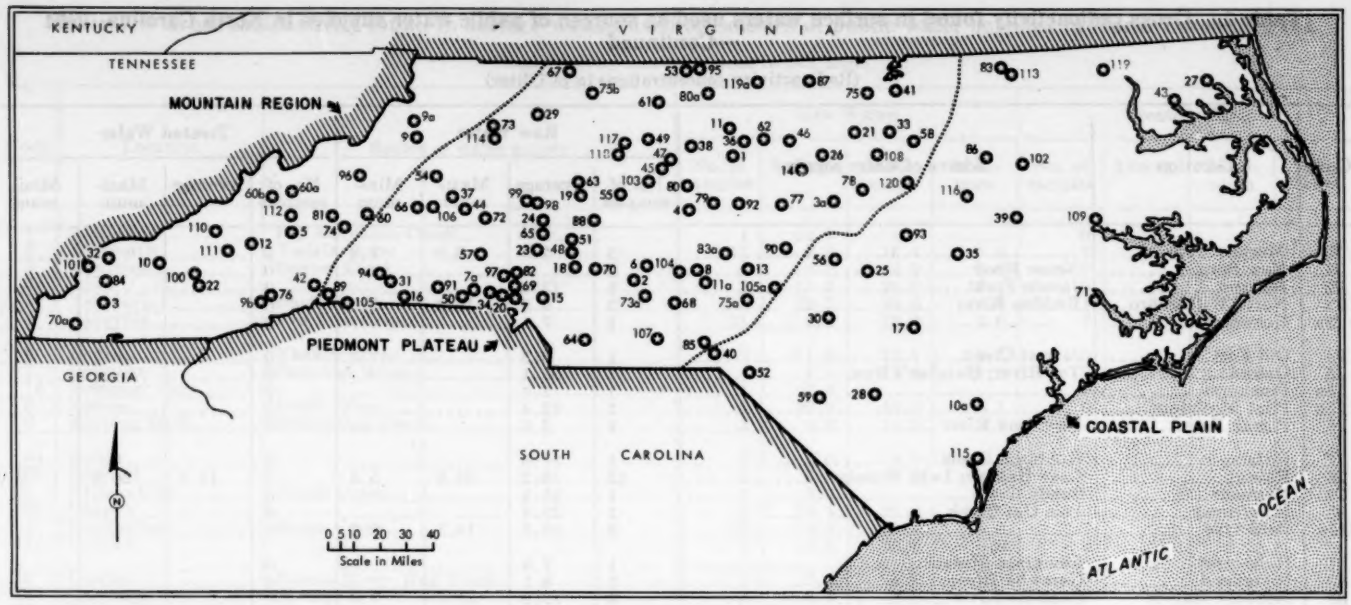

Figure 1. North Carolina surface water sampling stations with code numbers

the first runoff and then allow the balance of the rainwater to pass into the cistern. A 1-liter sample is usually collected monthly and analyzed for gross radioactivity. Table 2 presents the results for five cistern water supplies on Ocracoke Island and in Durham County, from April through December 1964.

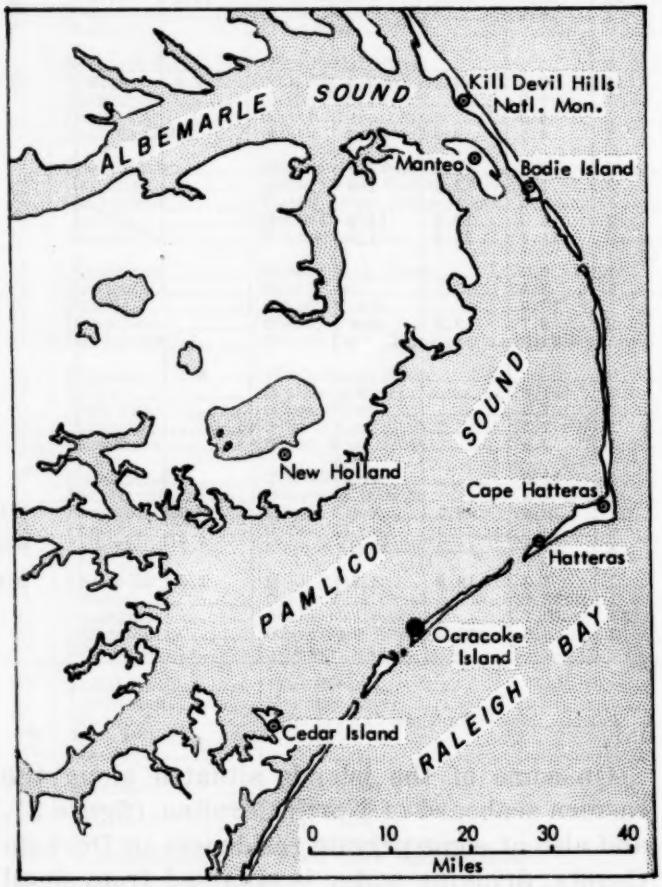

Figure 2. Eastern North Carolina, showing location of Ocracoke Island

Table 2. Gross radioactivity in cistern water in North Carolina, 1964

Location	Radioactivity concentrations, pCi/liter			
	Number of samples	Average	Maximum	Minimum
Ocracoke Island:				
Marina	7	67.2	143.1	17.5
Coast Guard Station	8	70.5	127.4	21.8
Residence A	1	80.1	80.1	80.1
Durham County:				
Residence B	6	67.4	85.7	27.9
Residence C	2	118.6	118.8	118.3

Discussion and interpretation of data

All results may be interpreted on the basis of the Public Health Service Drinking Water Standards (*4*), which state that in the absence of strontium–90 and alpha emitters, a water supply is acceptable if the gross beta activity does not exceed 1,000 picocuries per liter of water. This value relates to average intake from water over a long period of time and not from transient conditions.

The results for 1964 ior ground water, raw surface water, finished water, and cistern water has been summarized in table 3, by region of the State (figure 1).

Table 3. Summary of results of gross radioactivity in water supplies in North Carolina, 1964

Type of water and region	No. of samples	Average	Maximum	Minimum
Ground water:				
Mountain region	10	2.5	5.2	0.7
Piedmont region	43	2.9	52.2	1.3
Coastal region	75	6.2	20.7	1.5
Raw surface water:				
Mountain region	54	4.4	13.9	1.3
Piedmont region	313	12.1	64.7	1.0
Coastal region	79	15.6	38.0	3.2
Finished water from surface supplies:				
Mountain region	7	5.7	12.3	2.4
Piedmont region	128	8.0	26.7	1.8
Coastal region	45	8.4	24.4	3.2
Cistern water:				
Ocracoke Island	17	69.6	143.1	17.5
Durham County	8	80.2	118.8	27.9

It is apparent that the coastal region has somewhat greater radioactivity in its ground water than the Piedmont and mountain regions, as indicated by averages of 6.2, 2.9, and 2.5 pCi/liter, respectively. These activities would be considered as background since the radioactivity present in the water is imparted to the ground waters from the dissolution of geological formations of granite, gneiss, and other bedrock. Radioactivity so imparted would be the combined contribution of such elements as natural uranium, thorium, radium, potassium and other long-lived materials occurring in nature.

It can be noted that the mountain surface waters contain less radioactive material (4.4 pCi/liter) than those in the Piedmont (12.1 pCi/liter) and coastal regions (15.6 pCi/liter). This is to be expected, since the surface waters in the mountains have fewer dissolved solids. As these waters flow toward the ocean their dissolved solid content increases, paralleled by increases in their natural as well as their fallout radioactivity concentrations.

The mountain region had an average of 5.7 pCi/liter in finished water, while the Piedmont and coastal regions reported averages of 8.0 and 8.4 pCi/liter, respectively. In order to directly compare the results for raw water and finished water, one should refer to the data for individual samples of raw and finished water collected at the same time (*5*). It appears that finished water had, on the average, lower concentrations than raw water.

Radioactivity in the raw water decreased from a 1963 average of 17.1 pCi/liter to the 1964 average of 11.8 pCi/liter, or approximately by one-third. Finished water levels of activity decreased from a 1963 activity of 9.7 pCi/liter to the 1964 level of 8.0 pCi/liter. This indicates a removal of radioactivity for 1964 of 33 percent compared to a 1963 value of 47 percent.

It is readily apparent that during 1964, cistern water contained more radioactivity than any of the other types of public water supplies sampled. The average values were 69.6 pCi/liter on Ocracoke Island and 80.2 pCi/liter in Durham County. These values more closely parallel the values observed in precipitation during 1964 than the values for ground and surface water.

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Previous coverage in *Radiological Health Data:*

Period	*Issue*
June–August 1962 (cistern water)	*February 1963*
June 1958–December 1961	*May 1963*
November 1961–December 1963	*September 1964*

RADIONUCLIDE ANALYSIS OF COAST GUARD WATER SUPPLIES JANUARY–DECEMBER 1964

U.S. Coast Guard [1] and Division of Radiological Health [2]

Beginning in October 1961, 1-gallon samples of water have been obtained from seven Coast Guard Loran Stations in Alaska (figure 1).

The prime source of potable water at the Adak Loran Station is Alpine Lake which is at an elevation of 1,000 feet. The lake is occasionally frozen over with little snow cover each year from December through April. Water is delivered from the lake as needed through a 2-mile pipe line to three 25,000-gallon concrete reservoirs. All water is chlorinated at the station before use.

The Attu Loran Station's prime source of potable water is Alout Creek Dam and reservoir. This water source is usually frozen over from November through April, with a frequent snow cover of 1 to 3 feet. A 1-mile pipe line feeds water by gravity to three 25,000-gallon concrete reservoirs at the station. The water is chlorinated before use.

The Biorka Loran Station's water comes from a lake that is often frozen over lightly during January to March. The water is fed by gravity through a 1-mile long insulated pipe to a pump house. From there, it is pumped another mile to a 25,000-gallon steel storage tank from which it is fed by gravity one-half mile to the station, filtered, and chlorinated.

The source of water for the Cape Sarichef Loran Station is a small dammed creek and reservoir. The reservoir is usually frozen over from December through April, with 1 to 3 feet of snow cover during that period. The water is pumped 1½ miles from the dam to a 25,000-gallon steel storage tank above ground, from which the water is fed by gravity to the station, filtered, and chlorinated.

The Sitkinak Loran Station obtains its water from a natural lake which is normally frozen over from December through April, with 3 to 12 inches of snow cover during that period.

[1] U.S. Department of the Treasury
[2] U.S. Public Health Service

Figure 1. Coast Guard Loran Station water sampling locations

The water is pumped from the lake through a 1,500-foot line to three 25,000-gallon concrete reservoirs at the station. The water is filtered and chlorinated as used.

Port Clarence Loran Station's source of water is two shallow, flatland lakes, which freeze solid to their depth of 1½ to 3 feet from November through May. A variable snow cover of 1 to 5 feet is normal during this period. The water is pumped beneath the ice and seasonal frost to the station's concrete cisterns of 225,000-gallon capacity. Normally the entire capacity of the station's tanks is topped off prior to December 1 and only a small amount of water is pumped from the lake area after that date, depending on the thickness of snow, ice, and frost cover during the winter months. At the time of the first thaw, usually in May, the station tanks are again filled and kept filled until the following winter freeze. All water is filtered and chlorinated before use.

A natural hillside lake is Spruce Cape Loran Station's source of water. The lake is generally ice-free throughout the entire winter season with only light snowfall and general slush. The water is pumped one-half mile from the lake to a 25,000-gallon wood-stave cistern from which it is fed by gravity to the station, filtered, and chlorinated.

Water samples are analyzed for gross alpha, gross beta, barium-lanthanum–140, cesium–137, iodine–131, ruthenium–103 — ruthenium–106, and zirconium-niobium–95. The radionuclide analyses are performed by gamma spectrometry. All analyses are performed by the Southwestern Radiological Health Laboratory, Las Vegas, Nevada. The results of the gross alpha and beta analyses are reported in table 1. Values for specific radionuclide content are not reported, since in all cases they were below the respective minimum detectable levels (cesium–137, 5 pCi/liter; all others, 10 pCi/liter).

Table 1. Radioactivity in water samples from U.S. Coast Guard Loran Stations, January 1964–December 1964

(Concentrations in pCi/liter)

Alaskan location	Collection date, 1964	Gross alpha	Gross beta
Adak	no sample		
Attu Island	January 11	<1.1	36
	February 7	<0.1	2
	April 3	<0.1	50
	May 1	<0.1	14
	June 9	<0.1	11
	July 7	2.0	7
	August 2	1.8	10
	September 1	0.7	12
	October 6	<0.1	13
Biorka Island	February 26	2.9	41
	November 23	----	31
	November 24	----	3
	November 30	----	28
	December 17	----	26
	December 29	----	26
	December 30	----	26
Cape Sarichef	January 22	3.9	5
	March 10	<0.1	1
	May 12	0.3	5
	July 7	3.3	3
	July 15	2.2	2
	October 20	----	6
	December 11	----	11
Port Clarence	January 18	<0.1	12
	February 11	<0.1	13
	March 9	<0.1	6
	April 9	0.5	20
	May 7	<0.1	11
	June 6	<0.1	9
	July 7	1.5	13
	August 6	<0.1	18
	September 12	<0.1	21
	October 17	----	11
	November 12	----	10
	December 12	----	14
Sitkinak Island	January 13	<0.1	23
	February 12	<0.1	18
	March 17	<0.1	19
	April 10	0.5	11
	May 13	<0.1	18
	June 12	5.4	15
	July 7	1.4	3
	August 11	<0.1	12
	September 13	0.9	12
	October 22	----	10
	November 17	----	14
	December 16	----	10
Spruce Cape	January 7	<0.1	43
	February 10	1.1	40
	March 10	<0.1	<1
	April 6	0.5	39
	May 5	0.6	18
	June 9	<0.1	13
	July 27	1.0	<1
	August 3	0.9	12
	September 10	<0.1	4
	October 14	----	10
	November 9	----	16

Previous coverage in *Radiological Health Data:*

Period	*Issue*
October–November 1959	*June 1960*
October–December 1959	*July 1960*
Fourth quarter 1959, and first and second quarters 1960	*August 1961*
October 1961–April 1962	*October 1962*
May–December 1962	*August 1963*
January–December 1963	*November 1964*

Section IV—Other Data

FALLOUT RADIONUCLIDES IN MILK, TOTAL DIET, AND HUMAN BONE COMPARED TO FEDERAL RADIATION COUNCIL ESTIMATES, 1963-1964

Richard D. Grundy, Robert P. Chandler, James R. Coleman, and James G. Terrill, Jr.[1]

Estimates of the levels of fallout radionuclides in environmental media and human bone expected in 1963, 1964, and 1965 from the atmospheric testing of nuclear weapons conducted through 1962 have been made by the Federal Radiation Council (*1,2*). Observed values for 1963 and 1964 based on surveillance measurements are presented and compared with these predictions. Discussion is centered on those ingested fallout radionuclides most significantly contributing to the internal radiation dose to man: strontium–89, strontium–90, iodine–131, and cesium–137.

Although the total diet is the most direct measure of intake of radionuclides, specific indicator foods are also important. Milk is both the single food item most often used as an indicator of the population's intake of radionuclides from the environment as well as the most rapid vector, other than air and water, by which man can be exposed. Fresh milk is consumed by a large segment of the United States population and contains most of the radionuclides occurring in the environment which have been identified as biologically significant.

Milk

Radionuclide concentrations in milk are closely monitored by the Public Health Service's Pasteurized Milk Network (PMN) at 63 sampling locations in selected United States municipalities (*3*). Additional information on radionuclide concentrations in milk are obtained through the Atomic Energy Commission's Tri-City Diet Study (*4*) and various State milk sampling programs.

Strontium–89. Following the resumption of atmospheric nuclear testing in late 1961, an influx of strontium–89 into milk was observed by the PMN (*5*). The average monthly concentration showed a general increase and exhibited noticeable peaks in the springs of 1962 and 1963. The concentration of this radionuclide in milk declined after the spring of 1963, becoming negligible by the spring of 1964. The maximum annual average strontium–89 concentration at any stations during this period was 170 pCi/liter. The intake of radionuclides from the diet can be estimated assuming a 1-liter daily intake of milk. This would amount to 170 pCi/day.

Strontium–90. The observed strontium–90 concentrations in milk for the PMN and the Tri-City Diet Study from 1961 through 1964 are given in table 1. For consistency with the Federal Radiation Council presentation, these concentrations are presented on a "wet" or "dry" area basis (*2*), along with concentrations estimated by the Federal Radiation Council. It is observed that in the "wet" area, data produced by the two programs were in close agreement. In 1963, the PMN results were slightly below the Federal Radiation Council predictions.

In 1964, the results were slightly below the predicted concentration for both programs. Although not striking, the observed differences suggests the 1965 predictions were somewhat high, indicating that environmental contamination has not been as great as anticipated for the "wet" areas.

Considering the "dry" areas, the Tri-City Diet Study milk results were in agreement with Federal Radiation Council predicted concentrations, while PMN results were slightly higher than the predicted concentrations. The observed difference suggests the "dry" area predicted concentration for 1965 might be

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low, relative to the Pasteurized Milk Network results. This suggests that environmental contamination in the "dry" areas was somewhat greater than anticipated.

Iodine–131. Iodine–131 concentrations in milk for the PMN increased in 1961 with the resumption of atmospheric nuclear testing. However, as predicted, the iodine–131 had become undetectable by mid–1963 (*2*). The peak annual average was 104 pCi/liter at Palmer, Alaska, in 1962. Using the assumption of 1 liter daily intake of milk, this annual average would correspond to Range III (100–1,000 pCi/day) as defined by the FRC (*6*).

Cesium–137. Cesium–137 concentrations in milk for the Pasteurized Milk Network paralleled the strontium–90 results. A continued increase was observed from 1961 through 1964 when the peak concentration of cesium–137 in milk occurred. The annual averages in 1964 were 119 pCi/liter in the "wet" area and 86 pCi/liter in the "dry" area.

Diet

Estimates of children and teenager radionuclide intakes have been obtained since 1961, through the Institutional Total Diet Sampling Network (*7*) of the Division of Radiological Health and the Division of Environmental Engineering and Food Protection. This network, along with the Atomic Energy Commission's Tri-City Diet Study (*4*), the Food and Drug Administration's Teenage Diet Survey (*8*), the State of California's Diet Study (*9*), and the State of Connecticut's Standard Diet Study (*10*), provides nationwide estimates of dietary intakes of radionuclides. The characterization of radionuclide concentrations in the diet constitutes an important element of any integrated program of population-oriented radiation surveillance.

Strontium–89. The influx of strontium–89 into the diet was observed in the Institutional Diet samples in late 1961 following the resumption of atmospheric nuclear testing (*11*). The average daily intake was seen to reach peaks in mid–1960 and 1963, declining thereafter (*9*). This radionuclide had diminished to negligible levels by early 1964. The annual average daily intake was approximately the same in mid-1962 and mid-1963. These observations were in agreement with the Federal Radiation Council Report Number 4. The maximum annual average strontium–89 intake at any institution during this period was 182 pCi/day.

Strontium–90. Unlike strontium–89, strontium–90 has been observed in the Institutional Diet samples continuously since the inception of the network. During 1961, the network annual average strontium–90 intake was 8 pCi/day, which corresponds to approximately 7 pCi/g calcium in the diet.[2] After the resumption of nuclear testing, the average daily intake increased steadily, reaching a maximum in the spring of 1964. The network annual averages were 13 pCi/day in 1962; 25 pCi/day in 1963; and 32 pCi/day in 1964, corresponding to approximately 12, 22, and 28 pCi/g calcium, respectively. The highest annual average for an individual institution, 48 pCi/day, was observed during 1964. This corresponds to Range II (20–200 pCi/day) as defined by the Federal Radiation Council (*6*). A decreasing trend, first observed in the latter part of 1964, should be expected to continue through 1965.

Similar patterns of strontium–90 dietary intake were reflected in the Tri-City Diet Study (*8*) and the Teenage Diet Survey (*4*). Strontium–90 dietary intakes increased during 1961 and 1962, reaching a maximum in the spring of 1964.

Comparisons of these dietary results with the Federal Radiation Council estimates (*1,2*) are shown in table 2. For the "wet" area, the results from the two sampling programs were in reasonable agreement for all years, although the ratios of pCi/g calcium observed were below the values predicted by the Federal Radiation Council. In the "dry" area the observed results for both programs were also somewhat lower than the predicted ratio. However, in 1964, the Institutional Total Diet Sampling Network result, representing several sampling locations, was in closer agreement with the Federal Radiation Council predictions than the Tri-City results, which are represented by San Francisco, California.

[2] Based upon 1.13 g calcium per day (*11*).

Iodine–131. When considering the daily intake of iodine–131, emphasis is generally placed on the contribution from fresh milk, since the normal delay between deposition and the consumption of most other dietary components provides time for significant reduction of iodine–131 by decay. The dietary intake of iodine–131 as observed in the ITDSN reached a peak in 1961, with an average annual intake of 27 pCi/day. A decrease was seen in 1962, 1963, and 1964, with annual averages being 25, 16, and 0 pCi/day, respectively. Consistent with the predictions of the FRC (*2*), iodine–131 was no longer observable in the diet by mid–1963.

Cesium–137. During 1961, the cesium–137 dietary intake, as observed in the Institutional Total Diet Sampling Network, averaged 29 pCi/day. Subsequently, the dietary intake of cesium–137 increased, paralleling the strontium–90 intake. A peak network average daily intake of 154 pCi/day was observed in 1964. This is well below the value of 4,400 pCi/day for the population at large which can be derived from International Commission on Radiological Protection recommendations (*12*). Federal Radiation Council estimates of dietary intake of cesium–137 have not been made. However, as with strontium–90, cesium–137 dietary intakes are expected to decrease in 1965. Results of the Teenage Diet Survey further substantiate this expected decrease (*8*).

Bone

Indications of the extent to which strontium–90 has entered the food chain of man have been shown by the preceding summary of results. The resultant levels of strontium–90 in man can be described by employing the ratio of pCi ^{90}Sr/g Ca in bone.

For consistency with the presentation in Federal Radiation Council Reports Numbers 4 and 6, the 0-to 4-year old age group is considered. Table 3 shows the observed pCi ^{90}Sr/g Ca in bone from AEC's Health and Safety Laboratory (HASL) and Public Health Service programs along with Federal Radiation Council estimates. Reasonable agreement is noted between the predictions and the observed data. However, the observed values are consistently about one-half the predicted values. A difference of this order is not unexpected since the predicted values are for new bone being formed, whereas a considerable fraction of the bones of individuals 0–4 years of age was old bone, which (in 1963 and 1964) had a lower strontium–90/calcium ratio than the bone most recently formed.

Estimated dose

From the observed strontium–90 in bone and cesium–137 concentrations in milk it is possible to estimate annual dose rates to children. The resultant radiation dose rate from the indicated strontium–90 concentrations in bone may be estimated from the dose conversion factor presented by the Federal Radiation Council (*13*). The highest average bone level shown (6 pCi/g Ca), would result in a dose to bone marrow of approximately 5.5 mrad in one year.

For cesium–137, the yearly dose (assuming equilibrium) can be calculated from the following:

$$D = (11)\frac{I}{W}1.44T_e$$

Where:

D = dose rate (rads/year)
I = daily intake of cesium-137 (μCi/day)
W = body weight (kg)
T_e = effective half-life (days)

For infants the effective half-life is taken to be 30 days and the body weight as 10 kg (*13*); also, it is assumed that 1 liter is the daily intake of milk. Considering the "wet" area with a 1964 average for cesium–137 of 119 pCi/liter of milk, the resultant dose would be approximately 5.7 mrad for the year.

Since both nuclides contribute to bone-marrow dose, the sum of the estimated doses is of significance (*13*). Thus, the estimated total dose to bone marrow from strontium–90 and cesium–137 would be approximately 10 mrad during 1964 for young children in the "wet" area.

For long term low-level situations, such as represented by these data, a Protective Action Guide has not been recommended by the Federal Radiation Council. Rather, annual doses from fallout equal to or greater than the numerical values of the Radiation Protection Guides (RPG's) can be used as an indication of when there is a need for careful evaluation of fallout exposures. The RPG for bone marrow for the general population is given as 170 mrad/year (*6*). Thus, in the above case, the dose rate due to strontium–90 and cesium–137 for young children in the "wet" area amounts to approximately 6 percent of the RPG.

Table 1. Annual averages of strontium-90 content of U.S. milk, 1961–1964, compared with Federal Radiation Council estimates for 1963–1965

(pCi/g Ca)

Year	"Wet area"			"Dry area"		
	New York City (Tri-City)	FRC Estimate	PMN	San Francisco (Tri-City)	FRC Estimate	PMN
1961	10		10	5		5
1962	15		15	5		10
1963	30	[a]30	25	10	[a]10	15
1964	25	[b]30	25	10	[b]10	15
1965		[b]25			[b]5	

[a] Estimated in Federal Radiation Council Report No. 4.
[b] Estimated in Federal Radiation Council Report No. 6.

Table 2. Annual averages of strontium-90 content of U.S. total diet, 1961–1964, compared with Federal Radiation Council estimates for 1963–1965

(pCi/g Ca)

Year	"Wet area"			"Dry area"		
	New York City (Tri-City)	FRC Estimate	Institutional Diet	San Francisco (Tri-City)	FRC Estimate	Institutional Diet
1961	5		10	5		5
1962	10		10	5		10
1963	30	[a]50	25	10	[a]35	15
1964	30	[b]40	30	10	[b]20	20
1965		[b]30			[b]15	

[a] Estimated in Federal Radiation Council Report No. 4.
[b] Estimated in Federal Radiation Council Report No. 6.

Table 3. Average strontium-90 content of human bone in the United States, for 0–4-year olds at death, 1958–1965

(pCi/g Ca)

Year	"Wet" area			"Dry" area		
	Estimated [a]	Observed		Estimated [a]	Observed	
		HASL	PHS		HASL	PHS
1958		2.0			2.0	
1959		2.7			2.2	
1960		2.4			1.8	
1961		2.6			0.9	
1962		3.1	4.0		1.1	
1963	12	5.0	4.2	9	1.9	
1964	10	6.1	[b]6.1	5	2.6	2.7
1965	8			4		

[a] 1963, 1964, and 1965 values are predicted by assuming one-fourth of the dietary level of strontium-90 is laid down in new bone in 0- to 4-year old children (*1*, *2*), and that the strontium-90/calcium ratio of old bone in this group is the same as that of new bone.
[b] Values based upon results for January through June 1964.

Summary

The estimates made in Federal Radiation Council Reports Number 4 and 6 regarding the maximum concentrations of radionuclides in the environment and man have been essentially verified by recent surveillance data. The peak intake of radionuclides by the population groups selected appears to have occurred in 1964. It is concluded, therefore, that the predictions made in Federal Radiation Council Reports Number 4 and 6 and the radiation doses projected over the next 30 and 70 years are valid and reasonable for the situation as it exists currently.

For the situation wherein the major nuclear powers are abiding by the atmospheric test ban treaty, current surveillance operations are appropriate. This is due in large measure to cooperation between Federal and State agencies in developing nationwide coordinated surveillance programs. Such related surveillance programs complement each other and frequently provide corroborative information. In general, current surveillance activities are designed to yield measurements of environmental radiation levels which can ultimately be translated to estimates of average doses to population groups. Based upon predictions of dietary contamination levels anticipated in 1963 and subsequent years, the Federal Radiation Council concluded in Reports Number 4 and 6 that health risks from radioactivity in food over the next several years are too small to justify protective actions to limit the intake of radionuclides by food, particularly milk and dairy products. The observed radionuclide levels are in agreement with these predictions, and therefore the conclusions of the Federal Radiation Council apply.

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RADIOACTIVE CERAMIC GLAZES

Leonard F. Menczer[1]

During the past several years articles have appeared in news media concerning radioactivity in certain types of glazed earthware (tableware). Since there is little in the literature on this subject, a brief review may be indicated, including additional information secured by the author.

Historical aspects

The use of uranium compounds to produce pigmented glazes for ceramicware such as platters, pitchers, vases, water mugs, coffee cups, and demitasses was reported as early as 1906 (*1*). The colors produced by using uranium source materials in the glazing process range from brilliant orange-red to lemon-yellow. At low firing temperatures, and in a high lead glaze, uranium pigments produce a red color, while in an alkaline glaze they develop a yellow color. The forms in which uranium is usually used are black oxide of uranium (UO_2) and sodium uranate (Na_2UO_4) (*2*). The radioactivity of such glazed materials has been recognized for some time, and their use as sources for classroom demonstrations of radioactivity has been suggested (*3*). This use of ceramicware, as well as normal use as eating utensils, has been described as unlikely to present a health hazard (*4*).

The use of uranium source materials for ceramic glazes was interrupted in 1943, when uranium ores became unavailable except for atomic development. Just how large a stock of uranium pigment materials was on hand at the several pottery manufacturing plants in the United States in 1943 when controls were placed on uranium ores, and just how long that supply lasted is difficult to determine. It is believed that, due to the lack of uranium ore supply, by 1944–1945 the use of uranium pigments in ceramic glazes was discontinued.

It is reasonable to assume that millions of such dishes and drinking ware were manufactured and sold prior to 1944–1945 and that many of these continue in use today.

It was not until about 1959 that uranium oxide was again commercially available. Since then, however, it has been used under Atomic Energy Commission regulation, which imposes conditions concerning the safety of the manufacturing process and limits the quantities of uranium in the final product (*5*). Excerpts from current regulations governing the quantity of uranium which can be used in ceramic glazes are set forth below:

"Exemptions

"40.13 Unimportant quantities of source material

"(c) Any person is exempt from the regulation in this part and from the requirements for a license set forth in section 62 of the Act to the extent that such person receives, possesses, uses, transfers, or imports into the United States:

" . . . (2) Source material contained in the following products: (i) glazed ceramic tableware, provided that the glaze contains not more than 20 percent by weight source material; (ii) glassware, glass enamel, and glass enamel frit containing not more than 10 percent by weight source material; but not including commercially manufactured glass brick, pane glass, ceramic tile or other glass, glass enamel, or ceramic used in construction . . ." (*6*).

To clarify the term "Source Material" the following definition is used by the Commission:

"40.4 Definitions

"(h) "Source Material" means (1) uranium or thorium, or any combination thereof, in any physical or chemical form or (2) ores which contain by weight one-twentieth of 1 percent (0.05 percent) or more of (i) uranium, (ii) thorium or (iii) any combination thereof. Source Material does not include special nuclear material" (*6*).

The clause "not more than 20 percent by weight source material" developed "through expression from various manufacturers as to the quantities of source material necessary to give desired colors in the glaze. This quantity of source material in the glazed ceramic tableware was evaluated by the AEC staff from the radiological safety standpoint, taking into ac-

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count such considerations as the method of incorporating the radioactive material into the glaze, the type of radiation involved, the radiation levels at varying distances from the glazed ceramic tableware, the conditions of use of the tableware, and the parts of the body likely to receive radiation exposure in connection with the use of the tableware. The radiation safety evaluation made indicated that the radiation exposure to persons possessing and using glazed ceramic tableware containing not more than 20 percent source material in the glaze would be insignificant" (*7*).

More recently (March 1965), the AEC published criteria for the approval of products intended for use by the general public, containing byproduct material and source material (*8*). Essentially, the principal considerations in these criteria are similar to those mentioned above in evaluating glazed ceramic tableware.

AEC estimates of exposure from handling such tableware have been made on the basis that the highest exposure would be to the hands. The AEC has indicated that even if it is postulated, as an example of an extreme period of exposure, that there is a continuous contact with the tableware for as long as 1½ hours per day, with a radiation level of 5 mrad per hour at contact, then these conditions would produce an exposure of 7.5 mrem per day to the hands, or 2.6 rems for a 50–week period. This estimated exposure may be compared with 75 rems permissible exposure to the hands for a 50–week period for workers in the atomic energy industry (*9*).

From the inquiries by the writer, it was found that at least five manufacturers of china and earthenware at one time or another prior to the mid-forties used glazing pigments containing uranium compounds. However, only one of these five manufacturers as of 1963 used this type of pigment. In more recent years, several additional companies have made application for licensure from AEC to use depleted uranium materials for glazes used in ceramic or glassware manufacture.

Reviewing the above background information, it would appear that the public health interest in this subject centers on quantitating information on a number of problems. One of these is to ascertain the radioactivity associated with uranium pigment-glazed ceramicware manufactured prior to 1944–1945. Another is to determine the radioactivity of ceramicware manufactured after 1949 with glazing pigments consisting of depleted uranium oxide. Finally, if warranted, actual conditions of use of ceramicware in the general population should be determined to establish more precisely the dosimetry involved. Information bearing on the first of these problems is presented below.

Findings

It is assumed that all earthenware in this study was manufactured prior to the 1940's. This assumption is based on the fact that the individuals from whom the earthenware was collected stated in effect: "We've had these dishes for the past 20 to 30 years."

As the first step in evaluating the radioactivity associated with a wide assortment of glazed earthenware (tableware) collected in 1963, film badges were placed in direct contact with the glazed surfaces of selected items for a 24-hour period. Badge holders were not used, cadmium and copper filters being taped to the badges. Table 1 lists the findings of gross beta and gamma radiation by color and type of earthenware, and location of badge. In some instances, double badges were employed in order to check on the consistency of the film badge readings.

Although a wide assortment of earthenware was submitted to a cursory survey using a CDV–700 Geiger-Mueller survey meter for the screening procedure, only selected colors and color combinations were found to be radioactive. For example, all orange-pigmented products of the five manufacturers contained varying amounts of radioactivity. One manufacturer's products were blended color combinations such as chocolate brown with orange speckling; green, orange, and blue; and one set, blue-green on the inside (not radioactive), orange on the outside (radioactive). Not all lemon-yellow items showed activity. Those that did were substantially less radioactive than the orange-pigmented products.

From a review of table 1, it will be noted that gross readings (beta plus gamma) for the orange earthenware tested range from a low of 3.8 mrad/hr to a high of 19.6 mrad/hr. When examined for their beta and gamma components, gamma readings ranged from zero to 7.5 mrad/hr. In several cases (badges 17, 19, and

Table 1. Film badge readings obtained from surface contact with ceramic glazes

Badge No.	Color	Type of earthenware	Location of badge	24-hour reading (mrad)				mrad/hr maximum gamma plus beta
				Minimum gamma	Maximum gamma	Beta	Total maximum plus beta	
1 & 2	Chocolate brown	Dinner dish round—scalloped edge	Center of plate	0 0	70 70	140 140	210 210	9 9
3 & 4	Orange	Same	Same	0 0	50 60	130 140	180 199	8 8
5 & 6	Orange	Dinner dish square—scalloped corner	Same	0 0	50 50	140 150	190 200	8 8
7 & 8	Orange	Dinner dish square—scalloped corner	Same	0 0	90 70	150 160	240 230	10 10
9 & 10	Orange	Dinner dish round	Same	0 3	40 40	90 90	130 130	5 5
11 & 12	Orange	Saucer round	Same	3 0	15 60	160 160	175 220	7 9
13 & 14	Orange	Cereal bowl	Same	27 15	140 180	330 280	470 460	20 19
15	Orange	Saucer round—scalloped edge	Same	0	85	160	245	10
16	Orange & blue	Coffee cup	Lip portion outside	0	70	160	230	10
17 18	Orange	Coffee cup	Lip portion inside Lip portion outside	0 0	80 50	210 110	290 160	12 7
19 20	Orange	Water mug	Lip portion inside Lip portion outside	0 0	80 50	260 160	340 210	14 9
21 22	Orange	Coffee cup	Lip portion inside Lip portion outside	6 0	110 70	240 190	350 260	15 11
23 & 24	Orange	3-sectional dinner plate	Center	0 0	50 40	90 110	140 150	6 6
25 26	Green, orange, & blue	Matching dish and bowl	Dish, edge orange Bowl, on orange base	0 0	20 90	70 280	91 370	4 15
27 & 28	Lemon yellow	Dinner dish	Center of plate	0 0	3 3	10 10	13 13	0.5 0.5

21), where film badges were taped to the inside surfaces of coffee cups and mugs, substantially higher readings were observed than from badges taped to the outside. This may be simply an effect of geometry.

Discussion

Film dosimetry presents several problems. One encountered in this study was inadequate definition of beta–gamma energies, such as those occurring at the surfaces of ceramic glazes. Hence, the accuracy of the beta and gamma components reported in table 1 may be questioned. In general, the 0.010-inch copper filter on the film badge is considered to be opaque to beta radiations; however, this may not be true for beta particles with energies greater than 0.7 MeV. In addition, some of the soft gamma components are difficult to distinguish from higher energy beta particles. For these reasons a range of the gamma components is expressed in table 1 under column headings "Minimum gamma" and "Maximum gamma."

It is of interest to compare the findings in table 1 with reports received from the Atomic Energy Commission and the Public Health Service for, presumably, currently produced earthenware. These reports were issued as a result of inquiries from private individuals and official agencies following wide newspaper publicity in early 1963 in Connecticut. A communication from a representative of the AEC stated: "The beta radiation level on the dishes . . . is about the same as all dishes using uranium trioxide as a coloring agent. They read about 2 millirad per hour (2 mrad/hr) on contact . . . The amount of gamma or penetrating radiation which comes from the dishes is barely detectable with sensitive measuring instruments. It is barely distinguishable from natural background radiation" (*10*). A communication from the PHS states: "To be specific, their tests (the AEC) showed that about 14 percent 'source material' was used in the glaze for tangerine-colored dishes (which required more of the 'source material' in order to

attain the desired color than would be necessary for dishes with other colors), and with this concentration of 'source material' dose rate determinations at the surface of such dishes showed about 2 mrad/hr beta-gamma readings of about 0.5 mrad/hr gamma readings . . ." (*11*).

The average results reported by this study for dishes produced before 1944–1945 differ by approximately a factor of 5 from those reported by the AEC for dishes probably manufactured after 1949. However, the average ratio of beta-gamma to gamma absorbed dose observed in this study are reasonably consistent with the ratio determined from the AEC data (approximately 3.5 vs. 4, respectively). A number of possible explanations for these observed differences in activity are advanced:

a. Ceramic glazes produced during different periods (pre–1944–45 and post–1949) were being compared.
b. Sample geometries varied and/or different testing techniques were used.
c. The products of five manufacturers were investigated in this report while the AEC study may have been limited to the products of one manufacturer.
d. Data as to whether or not comparable glazes were being tested is lacking.
e. The quantity of glaze slurry employed may vary between samples.

Employing the values obtained in this survey, it is estimated that radiation exposure to the hands (using a 1½-hour daily period of exposure) ranges from a low of 2.0 rems per 50-week period to a high of 10.3 rems per 50-week period. The latter figure may be compared with the 75 rems allowable exposure to the hands of workers in the atomic energy industry. The 1½-hour period assumed for daily exposure to the hands would probably produce an "upper limit" of exposure; under conditions of normal use the duration of exposure would be shorter.

Summary

Utilizing the film badge method, gamma and beta-gamma radiation measurements were made at the surfaces of specific types of glazed ceramicware; these were compared with measurements reported by others. While the results were not strictly comparable, some of the differences observed may have been due to such variables as the geometry of the conditions under which the readings were taken and the thickness of the glaze. Also, the time periods (pre–1944–45 vs. post–1949) during which these glazed earthenware samples were produced may have been a factor.

The amount of exposure contributed by the glazed ceramicware discussed in this report is admittedly small; however, the documentation of any increment of artificial exposure, such as that discussed in this report, is of public health interest.

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ENVIRONMENTAL LEVELS OF RADIOACTIVITY AT ATOMIC ENERGY COMMISSION INSTALLATIONS

The U.S. Atomic Energy Commission receives from its contractors semiannual reports on the environmental levels of radioactivity in the vicinity of major Commission installations. The reports include data from routine monitoring programs where operations are of such a nature that plant perimeter surveys are required.

Summaries of the environmental radioactivity data for 22 AEC installations have appeared periodically in *RHD* since November 1960. Summaries follow for the Lawrence Radiation Laboratory, Los Almos Scientific Laboratory, and the Mound Laboratory.

Releases of radioactive materials from these installations for the periods covered in the reports below are governed by radiation protection standards set forth by AEC's Division of Operational Safety in directives published in the "AEC Manual." [1] These standards, which include radioactivity concentrations limits, are applicable to effluents released from AEC installations.

1. Lawrence Radiation Laboratory [2] July–December 1964

University of California
Berkeley, California

Berkeley Site

The Berkeley site of the Lawrence Radiation Laboratory (LRL) is located to the east of the University of California campus (figure 1). Technical facilities include a 6.3 Bev proton accelerator (Bevatron), a 700-MeV cyclotron, a 10-MeV linear accelerator, an 88-inch cyclotron, and various chemistry and physics laboratories.

The environmental sampling program includes monitoring of the atmosphere, surface and ground waters, sewage, and rain or dry deposition. Three types of atmospheric samples are taken: stack samples, local area samples, and perimeter samples.

Approximately 110 "stacks" with potential for releasing radioactive contaminants are sampled continuously. One-inch diameter filters are used at a flow rate of 1 liter per minute. The filters are changed weekly and counted for beta activity by an end-window Geiger-Mueller tube and for alpha activity by a thin-window proportional counter. Local area and perimeter air samples are taken at locations on the Laboratory site and at the property line, respectively. The samples are taken on 4- x 9-inch HV-70 filter paper at 4 cfm. The filters are changed weekly, counted for alpha activity by a thin-window proportional counter, and for beta activity by a 30 mg/cm ^{2}-window Geiger-Mueller tube. A 40-percent loss in the alpha count is assumed for self absorption. The average levels of activity observed in each type of sample are given in table 1.

Rain or deposition samples are collected monthly in 18-inch diameter cylindrical vessels lined with polyethylene bags at local area and

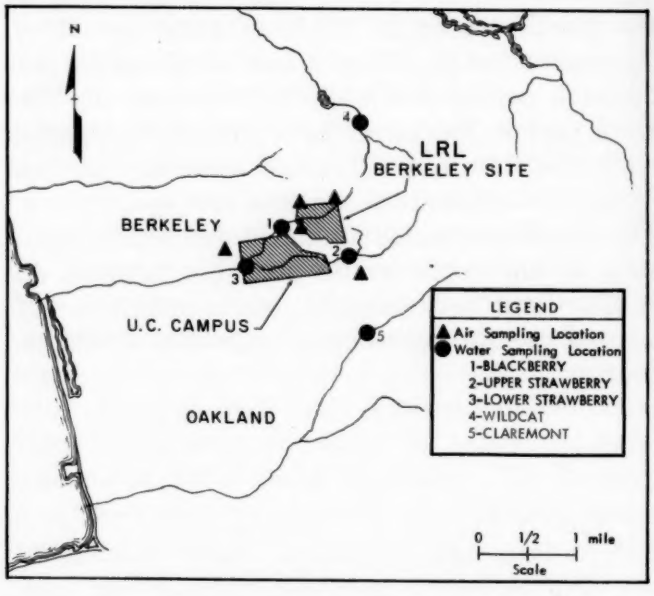

Figure 1. Environmental sampling locations at the Berkeley site

[1] Part 20, "Standards for Protection against Radiation," AEC Rules and Regulations, contains essentially the standards published in the "AEC Manual." The AEC Rules and Regulations are available from the Superintendent of Documents, U.S. Government Printing Office, Washington, D.C. 20402, on a subscription basis at $3.50 for 3 years.

[2] Summarized from "Lawrence Radiation Laboratory Environmental Radiation Levels—Berkeley and Livermore, Second Half 1964 and Annual Summary 1964."

Table 1. Atmospheric monitoring, Berkeley site July–December 1964

Sampling locations	Number of samples	Alpha (pCi/m³)	Beta (pCi/m³)
Stacks	4,000	0.01	1.24
Local area	249	0.00	0.46
Perimeter	98	0.01	0.55

perimeter sites. Rain samples are poured out and evaporated in beakers. If the bag is dry, it is rinsed out with dilute nitric acid, and the resulting solution is then evaporated. Final evaporation is conducted in 2-inch diameter stainless steel planchets, which are then flamed and coated with a thin film of lacquer. These planchets are counted for alpha activity in an internal-flow proportional counter and for beta activity with a thin-window, low-background Geiger-Mueller flow counter. No correction is made for self-absorption in the sample. Deposition data are given in table 2.

Table 2. Total deposition, Berkeley site, July–December 1964

Location	Number of samples	Alpha (nCi/m²)	Beta (nCi/m²)
Local area	60	0.0734	13.51
Perimeter	24	0.1428	13.89

Water samples are taken from sewers, onsite streams, and offsite streams. Two sewer lines serve the LRL area. The Hearst sewer receives waste from the larger part of the area. A sampling system takes a continuous proportional sample from the Hearst sewer as it leaves the Laboratory boundary. Samples are also taken of acid waste feeding into the Hearst sewer from Buildings 70, 70A, and 71. The Strawberry sewer receives waste from the southeast part of the Laboratory site. Building 74 is the most likely contributor of radioactivity to this sewer line. The two acid waste systems from Building 74 are sampled individually on a continuous basis. Strawberry and Blackberry Creeks comprise the Laboratory's storm drainage. These are sampled weekly at three locations. Two other nearby offsite creeks are also sampled weekly. All water samples are handled in the same manner as rain samples. The results from the water sampling program are presented in table 3.

Table 3. Water monitoring, Berkeley site July–December 1964

Type of sample	Number of samples	Alpha (pCi/liter)	Beta (pCi/liter)
Sewage:			
Hearst sewer	81	3.75	32.0
Strawberry sewer	41	<.4	66.7
Tap water	23	.06	5.5
Surface water:			
Onsite streams	78	.60	11.0
Offsite streams	51	.49	4.3

Livermore Site

The Livermore site of LRL is located about 50 miles southeast of San Francisco, California (see figure 2). Technical facilities include a small cyclotron, a 2-megawatt swimming pool reactor, and physics and chemistry programs associated with a weapons development program.

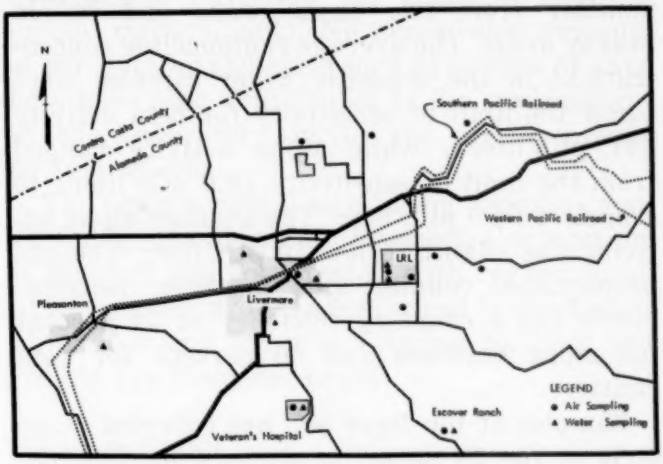

Figure 2. Sampling locations at the Livermore site

An environmental sampling program is maintained to provide information regarding the effectiveness of control measures and to determine whether any radiological changes in the environment are the result of laboratory operations. The sampling program includes air particulates, soil, domestic water, sewer effluent, and sewage plant products. Air samples are collected to ascertain that control efforts are restricting the release of radioactivity from the Laboratory to levels which do not exceed the permissible levels for the neighborhood around an atomic energy facility. The water samples are collected to monitor radioactivity in an underground water supply which provides most of the domestic water for the cities of Livermore and Pleasanton, and the sole supply for ranches in the Livermore and Amador Valleys.

Air samples are collected continuously at 15 sites within 5 miles of the Laboratory. Samples are collected at a rate of 4 cfm on 4- x 9-inch HV–70 filter papers, which are changed after every 7 days of operation. A minimum decay period of 96 hours is observed before the samples are counted, to eliminate the effect of natural radon and thoron daughters. The alpha activity remaining is measured with a counter equipped with a zinc-sulfide scintillation detector with a counting efficiency of about 24 percent for 5.1 MeV. The beta activity is measured with a Geiger-Mueller tube using strontium–90 in equilibrium with yttrium–90 as the standard. Alpha activity in air samples averaged 0.003 pCi/m^3, while beta activity averaged 0.23 pCi/m^3.

Domestic water samples are collected monthly from two onsite wells and in six nearby areas. The average radionuclide concentrations in the domestic water samples were below the limit of sensitivity for beta activity (41 pCi/liter), while alpha activity ranged from the limit of sensitivity (8.9 pCi/liter) to a high of 58.0 pCi/liter. The average alpha activity was slightly below 10 pCi/liter. The gas proportional counter used in these measurements has a counting efficiency of 34 percent for alpha emitters and 50 percent for beta emitters.

Samples of top layer soil are collected quarterly at the 19 sampling stations surrounding the Livermore site. The alpha activity fluctuated from the limit of sensitivity (5.0 pCi/g) to 13.0 pCi/g. The beta activity fluctuated from the limit of sensitivity (23.0 pCi/g) to 68.0 pCi/g.

Samples are collected every Monday, Wednesday, and Friday at the sewer line leaving the southwest project boundary, where it connects with the Livermore domestic sewer line. Grab samples are collected monthly at the Livermore Sewage Disposal Plant to assure that the liquid effluent from the laboratory is not creating abnormal radioactivity concentrations either in the oxidation ponds (which overflow into a natural waterway) or in the dried sludge (which is used as an agricultural soil conditioner). Radioactivity levels in sewage oxidation ponds and dried sludge are summarized in table 4.

Table 4. Environmental sampling, Livermore, July–December 1964

(Average concentrations)

Type of sample	Alpha (pCi/liter)	Beta (pCi/liter)
Sewage	92	44
Oxidation ponds	15	50
Dried sludge	34	27

Site 300

Site 300 is located in a very sparsely populated ranching area about 17 miles southeast of the Lawrence Radiation Laboratory at Livermore. Air and water samples are taken to determine whether operations at Site 300 are changing the normal radioactivity levels in the vicinity. The seven air samplers at Site 300 are operated at about 50 cfm on a continuous basis, but the filter papers are changed on an irregular schedule. Most of these air samplers are located within the boundaries of the test site due to unavailability of power facilities offsite. Water samples are taken from six onsite wells because they are the only readily accessible source of underground water. No samples are collected from streams since there is no flow during the summer and fall months and only sporadic flow in the winter.

All air and water samples are processed at the Laboratory in Livermore. Soil samples are collected monthly at seven offsite locations. Only top layer soil is collected to determine fallout concentrations. The average radioactivity levels in all types of samples are summarized in table 5.

Table 5. Environmental sampling, Site 300, July–December 1964

(Average concentrations)

Type of sample	Alpha	Beta
Air (pCi/m^3)	0.0045	0.29
Soil (pCi/g)	7	24
Onsite wells (pCi/liter)	8.9–30	41–46

Recent coverage in *Radiological Health Data:*

Period	*Issue*
Second half 1961	*October 1962*
Calendar year 1962	*October 1963*
Calendar year 1963	*October 1964*
First half 1964	*May 1965*

2. Los Alamos Scientific Laboratory[3] Calendar Year 1964

University of California
Los Alamos, New Mexico

As part of the environmental monitoring program at Los Alamos, measurements of air particulate radioactivity, activity in precipitation, and gamma background are made periodically. The samples have been taken on the roof of building TA–50 (about 1¾ miles southeast of the Administrative Building, the former location) starting in March 1963. Gamma-background surveys were discontinued in March 1963.

Air monitoring

Airborne radioactive particulate matter is collected on 4-inch-diameter filters. The sampling rate was reduced from 46 m^3/hr in 1962 to 25.5 m^3/hr in 1963, due to the addition of an activated charcoal filter behind the first filter. Air samples are ordinarily collected for 24-hour intervals during the work week; 72-hours weekend samples are collected.

The filters are counted for beta activity 7 days after collection in a thin-window (methane) flow proportional counter, with an overall efficiency for ^{90}Sr–^{90}Y of 50 percent. During the year, air samples averaged 0.69 pCi/m^3 for beta activity, with a maximum of 3.75 pCi/m^3 occurring on October 26, 1964.

Precipitation monitoring

Collection is made in a 0.4 square meter rain collector which delivers 1 liter of water for each 0.1 inch of precipitation. It has been found that this arrangement collected activity even during relatively dry periods. By washing down the sides of the collector with 1 liter of distilled water, a suitable sample is obtained. These "wash" samples, as well as any precipitation, are reduced in volume, dry-plated on 1-inch stainless steel planchets, and counted in an automatic beta counting system, installed in late 1963. This system has a gas-flow proportional counter, which provides increased efficiencies and lower background. (Counting was previously done in an end-window Geiger-Mueller counter with a window thickness of 1.9 mg/cm^2.) During the year, daily deposition samples averaged 236 pCi/m^2 for beta activity, with a maximum of 10,894 pCi/m^2 occurring on April 29, 1964.

Previous coverage in *Radiological Health Data:*

Period	*Issue*
1960–1961	*October 1962*
1962–1963	*October 1964*

[3] Summarized from "Beta-Gamma Radioactivity in Environmental Air at Los Alamos, New Mexico, for 1964" (LA–3245–MS).

3. Mound Laboratory[4] July–December 1964

Monsanto Research Corporation
Miamisburg, Ohio

Of the radionuclides in use at Mound Laboratory, only polonium–210, plutonium–239, and hydrogen–3 (tritium) are potential environmental contaminants. No measurable amounts of penetrating radiation such as gamma or hard beta have been contributed to the environment by the Laboratory during the period covered by this report.

The environmental monitoring program, conducted by the Monsanto Research Corporation, is planned and coordinated with regard to all of the various projects performed in the Laboratory. Air and water monitoring methods and results are discussed below.

Air monitoring

A continuous air monitor for measurement of tritium, and particulate air sampling equipment for measurement of alpha activity, are mounted on a 1-ton panel truck for use in the routine monitoring of the environmental air at 95 locations selected within a radius of 20 miles. The choice of sites on a given day depends on the wind direction at the time of collection.

During the 6-month period ending December 1964, 298 samples were taken for determination of tritium in air. In all cases tritium was nondetectable.

Monitoring for possible polonium–210 and plutonium–239 released to the environment is accomplished by determination of long-lived gross alpha activity on filter paper samples.

[4] Summarized from "Environmental Monitoring Report: July-December 1964 and 1964 Summary" (MLM–1241).

Counting is done in a low-background proportional counter. Sufficient delay time is allowed after collection to minimize interference from the daughter products of radon and thoron. The measured concentrations of alpha activity in air are summarized in table 6. No specific determinations of polonium–210 or plutonium–239 were made, since the alpha activity remained substantially below the environmental MPC's for these nuclides.

Table 6. Long-lived alpha concentrations in air in the Mound Laboratory environment, July–December 1964

Direction from laboratory relative to wind	Number of samples	Concentration (pCi/m^3)
Upwind	54	0.0027
Downwind	323	0.0042

Water monitoring

Liquid radioactive waste materials from polonium operations at the Laboratory are processed in a special waste disposal plant designed to reduce radioactivity to a concentration level at which it may be discharged to the Great Miami River. Liquid waste from plutonium work is small in volume, is handled separately as a packaged waste, and is not discharged to the river. Helium–3, which is purified at the Mound Laboratory, contains small quantities of tritium. Liquid wastes from this work, also small in volume, are treated (diluted with water when necessary) to assure that the radioactivity content is below the maximum permissible concentration before discharge to the Great Miami River.

Weekly water samples are collected from a drainage ditch and five locations along the Great Miami River as shown in figure 3. The drainage ditch carries away all storm sewer water and liquid tritium wastes from the plant site. Sampling location number 2 is located at the point of discharge of the laboratory effluent to the Great Miami River, and number 6 is 5 miles downstream from the effluent outlet.

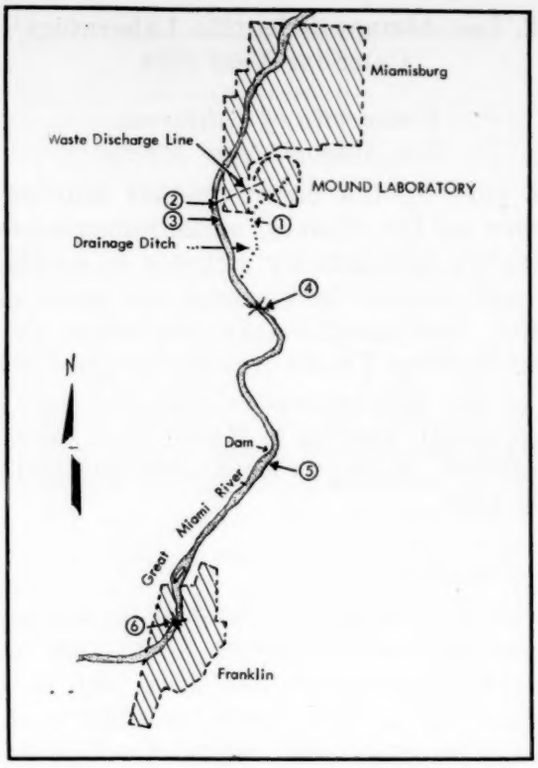

Figure 3. Water sampling locations in Great Miami River, Mound Laboratory

All of the river samples are analyzed for polonium–210. The drainage ditch samples and some of the river samples are analyzed for tritium. Average concentrations of tritium and polonium–210 are given in table 7.

Table 7. Offsite water monitoring for polonium-210 and tritium from Mound Laboratory, July–December 1964

Nuclide and station number (see figure 3)	Number of samples	Concentration (pCi/liter)
Polonium-210		
2 (effluent)	28	558
3 (250 yards downstream)	28	85
4	28	1.8
5	28	5.4
6	28	0.9
Hydrogen-3 (tritium)		
1 (drainage ditch)		1,000,000
2 (effluent)		0.0
4		0.0

Recent coverage in *Radiological Health Data:*

Period	*Issue*
January–June 1962	*March 1963*
July 1962–June 1963	*April 1964*
July–December 1963	*November 1964*
January–June 1964	*May 1965*

REPORTED NUCLEAR DETONATIONS, OCTOBER 1965

In early October 1965, the Atomic Energy Commission announced detection of seismic signals from a U.S.S.R. nuclear testing area. Later in the month, the Department of Defense announced a United States nuclear test, conducted by the Defense Atomic Support Agency.

Seismic signals recorded on October 8 indicated that a disturbance occurred in the Soviet Nuclear testing area in the Semipalatinsk region equivalent to a low to low-intermediate yield nuclear explosion. (Low is less than 20 kilotons of TNT equivalent; low-intermediate is 20–200 kilotons.)

The United States detonated an 80-kiloton thermonuclear device almost a half-mile underground on Amchitka Island, Alaska, on October 29, 1965. The purpose of this project was to obtain information on seismic signals from underground nuclear tests and their long-distance travel times. Such information may be useful in distinguishing between natural earthquakes and man-made nuclear detonations that are detected by seismographic equipment. The Division of Radiological Health participated in the project, providing offsite radiological health and safety support services, including the distribution of film badges, and sampling of air, water, milk, and vegetation. Following the event, all radiation monitoring instruments gave background readings only.